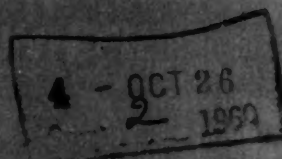


REACTOR CORE MATERIALS



A Quarterly Technical Progress Review

Prepared for U. S. ATOMIC ENERGY COMMISSION by BATTELLE MEMORIAL INSTITUTE

August 1960

● VOLUME 3

● NUMBER 3

TECHNICAL PROGRESS REVIEWS

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Reactor Core Materials (covering solid material developments), R. W. Dayton, E. M. Simons, and associates, Battelle Memorial Institute

Reactor Fuel Processing, Stephen Lawroski and associates, Chemical Engineering Division, Argonne National Laboratory

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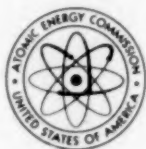
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REACTOR CORE MATERIALS

A REVIEW OF RECENT DEVELOPMENTS

Prepared for U. S. ATOMIC ENERGY COMMISSION
by BATTELLE MEMORIAL INSTITUTE



● AUG. 1960

● VOLUME 3

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foreword

Reactor Core Materials is prepared quarterly by staff members of Battelle Memorial Institute under the auspices of the Office of Technical Information of the U. S. Atomic Energy Commission. Information for each Review is drawn from all literature sources which have become available during the three-month period covered. In this way, *Reactor Core Materials* provides a ready reference to the highlights of the latest significant developments. A bibliography is provided at the end of each of the five sections for the benefit of readers who wish to obtain more detailed information in any particular area.

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REACTOR CORE MATERIALS

FUEL AND FERTILE MATERIALS

Two new books should be of interest to reactor metallurgists and designers. One of these books contains the papers delivered at the colloquium on the Effects of Radiation on Materials held at Johns Hopkins University in March 1957.¹ Included are discussions of current concepts of radiation effects and of results of experiments with fuel and structural metals and alloys, control and moderator materials, shielding materials, semiconductors, and organic substances. The other book contains the papers presented at the First International Symposium on Nuclear Fuel Elements held at Columbia University in January 1959,² and deals exclusively with nuclear fuel elements. Among the topics discussed are fuel elements of future reactors, standardization of elements, cost analysis for low-cost fabrication and reprocessing, and fuels processed from uranium, thorium, and plutonium. Also discussed are dispersion type fuels, high-temperature fuels, and the behavior of fuels under conditions of reactor operation. Detailed references are given in both books.

Unalloyed Uranium

The need for processing uranium ores for the U^{235} isotope is expected to continue to build the reserves of depleted uranium. Aside from its uses in shielding and as a fertile material, depleted uranium has no direct nuclear use. Consequently, two reports^{3,4} have been issued recently, stressing potential nonnuclear uses of this material. One of these³ presents a rather complete summary of uranium properties which is of common interest in both nuclear and non-nuclear fields.

Studies on the self-diffusion of uranium in the beta phase⁵ have been performed by means of couples of natural uranium and U^{234} -enriched uranium. The couples were hot pressed and

heat-treated. Penetration-concentration curves were obtained by alpha counting of sectioned layers. Diffusion coefficients determined from these curves are as follows:

$$5.1 \times 10^{-12} \text{ cm}^2/\text{sec at } 700^\circ\text{C}$$

$$7.9 \times 10^{-12} \text{ cm}^2/\text{sec at } 725^\circ\text{C}$$

$$1.1 \times 10^{-11} \text{ cm}^2/\text{sec at } 740^\circ\text{C}$$

$$1.6 \times 10^{-11} \text{ cm}^2/\text{sec at } 755^\circ\text{C}$$

Data obtained by Hanford⁶ on the effect of cyclic annealing on the tensile properties of irradiated uranium are presented in Table I-1. Also

Table I-1 EFFECTS OF CYCLIC HEAT-TREATMENT ON THE TENSILE PROPERTIES OF IRRADIATED URANIUM⁶

| Sample | Burnup, at. % | Annealing treatment* | Yield strength (0.2% offset), 1000 psi | Ultimate tensile strength, 1000 psi | Elongation, % |
|--------|---------------|----------------------|--|-------------------------------------|---------------|
| B-5 | 0.031 | 1 | 32.5 | 45.9 | 1.2 |
| M-2 | 0.075 | 1 | 22.4 | 33.5 | 2.5 |
| C-4 | 0.1 | 1 | 21.3 | 35.6 | 1.0 |
| M-6 | 0.075 | 2 | 30.5 | 43.7 | 1.0 |
| | 0.075 | 3 | 18.7 | 49.1 | 2.1 |
| C-3 | 0.1 | 3 | 22.4 | 30.5 | 0.9 |
| | 0.075 | 4 | 17.3 | 36.1 | 12.4 |
| | 0.1 | 4 | 29.1 | 32.2 | 4.6 |

*Annealing treatments: (1) Three alpha-to-beta cycles (six transformations) between 400 and 700°C; specimens were held for 2 hr at each terminal temperature. (2) Three alpha-to-gamma cycles (six transformations) between 400 and 800°C; specimens were held for 2 hr at each terminal temperature. (3) 700°C anneal for 10 hr. (4) 800°C anneal for 10 hr. All heating and cooling rates were 10°C/hr.

listed for reference are the tensile properties of similar specimens given a single pulse anneal. In general, very little increase in ductility occurred during repeated phase transformations. Sample M-6, which was cycled in the

gamma phase, had an elongation of 1.0 per cent; however, a similar specimen given a single pulse anneal had an elongation of 12.4 per cent. The yield strength for the 0.031, 0.075, and 0.1 at.% burnup specimens cycled between the alpha and beta phases decreased with burnup, contrary to the normal trend of radiation-damage recovery. All the specimens cyclicly annealed have exhibited an extensive crack network at the surface. The yield-strength behavior referred to above can be explained by considering the reduction in cross-sectional area caused by a crack network. The reduction in yield strength with burnup is consistent with the increased severity of cracking expected in higher burnup specimens.

The release of fission products from small cylindrical specimens of normal uranium heated in an air stream was also studied.⁷ These specimens were irradiated to 2.4×10^{14} nvt and heated at temperatures varying from 425 to 1440°C for 2.5 to 232 min. Xenon, iodine, and tellurium were the most volatile fission products. The maximum release from the specimen was 90 per cent for iodine and 80 per cent for xenon at the highest temperatures employed. The rate of release was proportional to the rate of uranium oxidation. Continued heating after all the uranium was converted to oxide did not release significant additional amounts of these elements. Cesium and ruthenium ranked next in order of volatility, with a maximum of 6 per cent of the cesium and 15 per cent of the ruthenium being released. The rate of release of these two elements was slightly time dependent, and ruthenium was released at a higher rate after all the uranium was oxidized. Strontium, barium, and zirconium were quite nonvolatile. In an air atmosphere, less than 0.2 wt.% release was observed for these elements.

Atomics International⁸ has shown that porous uranium compacts cycled between 100 and 500°C are perhaps only slightly more stable than dense uranium. Thermal-cycling tests from 200 to 700°C showed that the porous uranium was inferior to dense uranium. It was concluded that mechanical restraint would be necessary if this fuel were to be cycled into the beta phase.

The Nuclear Development Corporation^{9,10} has reported its evaluation of "sponge uranium" as a fuel for nuclear power reactors. The term "sponge uranium" is ascribed to metallic uranium powder that has been packed to a density of 70 per cent by volume and infiltrated with so-

dium. The aim of this concept is to produce fuel having the desirable characteristics of metallic uranium while avoiding its present limitations. The promise of this fuel, which lies in the possible relaxation of burnup and temperature limits encountered with metallic uranium, can be expected from (1) a shortening of the path of fission gases, (2) the ability to withstand gross distortion of the uranium particles without a reduction in the apparent fuel density, and (3) the ability of uranium particles to withstand cracking without affecting fuel-element performance. If these expectations are realized, sponge uranium should yield an economically attractive burnup affording appreciable savings in fuel-cycle costs over solid metallic fuels. Preirradiation evaluation of sponge uranium is being performed, and irradiation studies are being planned at 1100 to 1600°F and a burnup of 1 at.%.¹²

Papers of a review nature have been published on uranium oxidation¹¹ and the crystallographic mechanisms of deformation in alpha uranium.¹²

(M. S. Farkas)

Alpha-Uranium Alloys

In an investigation¹³ of the effects of irradiation on uranium-0.52 and 1.62 wt.% zirconium, wrought material, variously heat-treated, and castings were evaluated. Although the wrought and heat-treated alloys were reportedly stable under thermal cycling, it was found that they elongated rapidly under irradiation. The best material studied was the 1.62 wt.% zirconium alloy in the as-cast condition. It was found to increase in length initially at a rate of about 5 per cent per atomic per cent burnup, after which the rate steadily diminished. Up to burnups of at least 5.3 at.%, it retained a smooth surface. Two specimens of the cast 1.62 wt.% zirconium alloy, one of which was irradiated to 2.1 at.% burnup at a calculated central temperature of 690°C and the other irradiated to 5.3 at.% burnup at a calculated central temperature of 620°C, swelled and increased in volume with the formation of a large central void surrounded by highly porous metal. The critical swelling temperature of uranium-1.62 wt.% zirconium alloy was found to be between 300 and 620°C, and there were indications that it may be near 500°C.

The irradiation of clad and unclad extruded uranium-2 wt.% zirconium alloys was performed¹⁴ to determine an effective heat-

treatment for improved irradiation stability. Irradiation temperatures ranged from 200 to 750°C. For unclad specimens, it was found that the irradiation temperature strongly influenced the irradiation growth rates. Growth rates of the clad specimens were relatively insensitive to either irradiation temperature or prior heat-treatment, although cladding had only limited ability to restrain swelling at the more elevated temperatures. Results of this study indicate that the heat-treatment for the extruded uranium-2 wt.% alloy should consist of a gamma solution treatment at 800°C, followed by isothermal transformation at 690°C.

A visual study of the corrosion of defected Zircaloy-2-clad uranium and uranium-2 wt.% zirconium specimens has been reported by Battelle.¹⁵ Observations of the fuel specimens in high-purity pressurized water at 200 to 345°C were made by means of a windowed autoclave. Continuous measurement of the pressure increase, caused by the accumulation of hydrogen, served to monitor the progress of the reaction after clouding of the water by corrosion products made visual observation impossible.

The nature of the attack of all specimens was similar, although the time at which different stages occurred varied. Following an induction period, the first evidence of attack was the slow formation of a blister in the cladding area surrounding the defect. Eventually, a copious evolution of hydrogen occurred at the base of the swollen area. In general, a crack could be seen in the cladding at this stage. Catastrophic failure of the specimen followed swiftly. The time required for each phase of the reaction was reduced as the temperature was raised. Initial swelling occurred after about 24 min at 345°C, but required only 8 hr at 200°C. Of the materials selected for study, diffusion-treated uranium-2 wt.% zirconium specimens were most resistant to attack.

The mechanical and metallurgical properties of six high-uranium alloys were investigated by Atomics International.¹⁶ Tensile and creep properties were determined at 900°F. A uranium-3.5 wt.% molybdenum-0.1 wt.% aluminum alloy exhibited the best properties with an average ultimate strength of 110,000 psi and a 0.2 per cent offset yield strength of 83,000 psi. Following, in descending order of ultimate strength, were the uranium-3.5 wt.% molybdenum, uranium-3.5 wt.% molybdenum-0.1 wt.% silicon, and uranium-3.5 wt.% molybdenum-

0.5 wt.% aluminum alloys, and unalloyed uranium. With one exception, yield strengths were in the same order. The uranium-3.5 wt.% molybdenum-0.5 wt.% aluminum alloy possessed practically no ductility. Creep tests showed that 40,000 to 50,000 psi was required to attain secondary creep rates of greater than 0.01 per cent per hour for the uranium-3.5 wt.% molybdenum-0.1 wt.% aluminum alloy. This rate was attained at 20,000 psi for a uranium-3.5 wt.% molybdenum-0.5 wt.% silicon alloy and at 10,000 psi for a uranium-3.5 wt.% molybdenum alloy.

Thermal cycling between 200 and 1300°F showed that the uranium-3.5 wt.% molybdenum and uranium-3.5 wt.% molybdenum-base alloys were more dimensionally stable than the uranium and uranium-2 wt.% zirconium-0.1 wt.% aluminum specimens. Cycling tests between 200 and 900 to 1000°F gave similar results. Irradiation tests on aluminum-clad uranium-3.5 wt.% molybdenum and uranium-3.5 wt.% molybdenum-0.5 wt.% silicon alloys indicate that both can be considered dimensionally stable at a peak burnup of 3000 Mwd per metric ton of uranium.

Argonne¹⁷ has completed a study of uranium-fissium alloys. Hardness, density, and thermal-expansion data are presented for alloys of uranium with certain fission-product elements. The elements are those expected in a spent fuel from a fast reactor following pyrometallurgical refining. In cast and gamma-quenched alloys, retention of the high-temperature gamma phase produces low hardness and low density.

(M. S. Farkas)

Gamma-Phase Uranium Alloys

Hall effect and electrical-resistivity measurements of the metastable gamma phase in uranium-molybdenum and uranium-niobium alloys are reported by Atomics International.¹⁸ Alloys containing 21.7 and 30.5 at.% molybdenum and 22.2 at.% niobium exhibited positive Hall coefficients which were only weakly temperature dependent. The Hall coefficient for an 11.6 at.% molybdenum alloy was strongly dependent on both temperature and magnetic field. Below about 2°K, all samples were superconducting and exhibited slight negative temperature coefficients of resistivity from the transition temperature to room temperature. A magnetic field

Table I-2 GAMMA-PHASE DIFFUSION DATA IN THE URANIUM-TITANIUM SYSTEM¹⁹

| Uranium concentration, at.% | \bar{D} , cm ² /sec | | | | Q, kcal/gram atom | D_0 , cm ² /sec |
|-----------------------------------|----------------------------------|-----------------------|-----------------------|-----------------------|----------------------|---------------------------------|
| | 950°C | 1000°C | 1050°C | 1075°C | | |
| 5 | 1.7×10^{-9} | 2.6×10^{-9} | 4.5×10^{-9} | 5.7×10^{-9} | 30.2 | 4.6×10^{-4} |
| 10 | 1.0×10^{-9} | 1.6×10^{-9} | 3.1×10^{-9} | 3.4×10^{-9} | 33.8 | 1.1×10^{-3} |
| 20 | 4.6×10^{-10} | 7.1×10^{-10} | 1.3×10^{-9} | 1.9×10^{-9} | 37.5 | 2.2×10^{-3} |
| 30 | 2.3×10^{-10} | 5.7×10^{-10} | 8.9×10^{-10} | 1.1×10^{-9} | 39.4 | 2.6×10^{-3} |
| 40 | 2.7×10^{-10} | 4.2×10^{-10} | 8.6×10^{-10} | 1.2×10^{-9} | 39.4 | 2.6×10^{-3} |
| 50 | 2.9×10^{-10} | 4.6×10^{-10} | 1.1×10^{-9} | 1.5×10^{-9} | 42 | 9.5×10^{-3} |
| 60 | 6.2×10^{-10} | 8.6×10^{-10} | 1.7×10^{-9} | 2.6×10^{-9} | 38.4 | 4×10^{-3} |
| 70 | 1.1×10^{-9} | 1.5×10^{-9} | 3.1×10^{-9} | 4.1×10^{-9} | 34.8 | 1.6×10^{-3} |
| 80 | 1.9×10^{-9} | 2.8×10^{-9} | 5.5×10^{-9} | 6.5×10^{-9} | 33 | 1.4×10^{-3} |
| 90 | 3.4×10^{-9} | 5.4×10^{-9} | 1.2×10^{-8} | 1.25×10^{-8} | 36.6 | 1.1×10^{-2} |

of about 30 kilogauss was required to restore normal resistivity at 1.2°K. The relation between these results and the electronic structure of the gamma phase is discussed.

The coefficients for diffusion in the gamma phase of the uranium-titanium system are presented in a French report.¹⁹ The results are shown in Table I-2. Marker-movement meas-

at.% niobium at 800°C, from 60 to 65 at.% at 892°C, and from about 53 to 55 at.% at 996°C. The structure of the phase is not reported. Because of the number of previous studies of the system where such a phase was undetected, confirmation of the interpretation would be desirable.

Uranium-Molybdenum

A study of the U₂Mo phase in the uranium-molybdenum system is reported by the Fulmer Research Institute.²⁰ The limits of the region are in agreement with those reported by other investigators. Zirconium was soluble to about 2.5 at.% in the U₂Mo phase and delayed ordering considerably. Niobium also delayed the transformation, being soluble to the extent of 1.8 at.%. Chromium had little effect on the ordering kinetics.

The effects of tantalum, niobium, and rhenium additions on the kinetics of transformation of the gamma phase in the uranium-10 at.% molybdenum alloy were also investigated. Tantalum accelerated the transformation appreciably, whereas niobium, although it decreased the incubation time, had little effect on the subsequent rate of transformation. Rhenium delayed the transformation by factors of 2 to 5, depending upon the temperature of transformation. This delay is attributed to a decrease in free energy of the gamma phase as a result of the rhenium addition, which should exhibit a negative heat of solution in uranium-molybdenum alloys.

The French²² also report a study of the effect of ternary additions on the stability of the gamma phase in uranium-molybdenum alloys. Of the additions studied, which included chromium, niobium, rhenium, ruthenium, and zirco-

Table I-3 DIFFUSION IN THE URANIUM-TITANIUM SYSTEM BY MARKER-MOVEMENT MEASUREMENTS¹⁹

| | 950°C | 1000°C | 1050°C | 1075°C |
|---------------------------------|----------------------|----------------------|----------------------|----------------------|
| Uranium, at.% | 82 | 82 | 82 | 83.5 |
| Titanium, at.% | 18 | 18 | 18 | 16.5 |
| D_U , cm ² /sec | 4.7×10^{-9} | 9.5×10^{-9} | 1.6×10^{-8} | 2.2×10^{-8} |
| D_{Ti} , cm ² /sec | 1.2×10^{-9} | 2.9×10^{-9} | 4.1×10^{-9} | 5.8×10^{-9} |

urements lead to the data shown in Table I-3, where the concentration at the marker and calculated intrinsic diffusion coefficients are given.

Uranium-Niobium

A study of the uranium-niobium and uranium-niobium-zirconium systems is reported by Fulmer Research Institute, Ltd.²⁰ The results are in general agreement with those reported by earlier investigators with minor variations. The monotectoid is reported to occur at 640°C and 17.5 at.% niobium, the gamma miscibility gap at this temperature extending to between 65 and 70 at.% niobium. The peak temperature of the gap is given as 950°C at 50 at.% niobium.

The results of uranium-niobium diffusion couple analysis are reported by MIT.²¹ On the basis of the results obtained, a new and previously unreported phase is reported to exist in the system with a range of composition from 65 to 75

nium, ruthenium was found to be the most effective stabilizer.

(A. A. Bauer)

Epsilon-Phase Alloys

A report covering the preparation, irradiation, and irradiation test results of Zircaloy-2-clad U_3Si specimens has been issued by Chalk River.²⁴ Of four specimens irradiated to a maximum burnup of about 1600 Mwd per ton of uranium at 580 to 660°C, two specimens contained splits in the cladding, one axial and the other circumferential at the fuel-end cap interface. Although failure of the cladding in two of the specimens occurred, it appears that the irradiated U_3Si retains its corrosion resistance to water, based on the low fission-product activity in the irradiation loop prior to removal of the specimens. Diameter and volume changes were small at burnups in the range of 600 Mwd/ton. Swelling became appreciable as burnup increased; maximum volume changes of 7 per cent were obtained. It is concluded that the swelling of the fuel results from the agglomeration of fission gases and that the swelling temperature for U_3Si is below 500°C.

(A. A. Bauer)

Dilute Uranium Alloys

Aluminum-Uranium Alloys

Studies of the effects of ternary additions of tin and zirconium to the aluminum-35 wt.% uranium binary alloy were continued at Battelle.²⁵ Stress-rupture tests have been performed on ternaries containing 2 wt.% tin, 3 wt.% tin, 2 wt.% zirconium, and 3 wt.% zirconium in order to determine the stresses required for creep tests. The temperature for these tests was 250°C. The following data were accumulated:

| Alloy (balance Al), wt. % | Stress, psi | Time to failure, hr | Elongation, % | Reduction of area, % |
|---------------------------------|----------------|---------------------------|------------------|----------------------------|
| 35 U | 9500 | 227 | 8.9 | 14.3 |
| 35 U-2 Sn | 8500 | 57 | 9.8 | 14.4 |
| 35 U-3 Sn | 7000 | 130 | 11.4 | 18.0 |
| 35 U-2 Zr | 9000 | 92.5 | 17.1 | 24.2 |
| 35 U-3 Zr | 8500 | 48 | 13.8 | 7.9 |

These data offer a guide to the choice of proper stresses for relatively long-time creep tests. It is interesting to note that the binary alloy has substantially less ductility than the alloys with ternary additions.

Since one of the problems which might be encountered in bonding aluminum to nickel-plated uranium is the formation of the ternary eutectic, a study of the effect of naturally occurring impurities on the location of this eutectic temperature has been completed. A base alloy prepared from high-purity components exhibits a ternary eutectic temperature of $624 \pm 2^\circ C$. An alloy prepared from commercially available components exhibits a eutectic temperature of $621^\circ C$. It was found that the impurities most responsible for the depression of the eutectic temperature were iron and silicon, and these were effective only in the presence of nickel.

Zirconium-Uranium Alloys

Significant developments in the technology of zirconium-uranium alloys have been summarized in a report prepared by Battelle.²⁶ The latest phase diagram is presented and shows that at elevated temperatures the body-centered cubic forms of beta zirconium and gamma uranium are isomorphous. This body-centered solid solution decomposes at $595^\circ C$ ($1105^\circ F$) to form alpha zirconium and epsilon. The epsilon phase is stable up to $610^\circ C$ ($1130^\circ F$), where it forms peritectoidally from alpha uranium and body-centered cubic gamma solid solution. Oxygen has a marked effect on the constitution of the binary alloy. For example, a binary zirconium-35 wt.% uranium alloy is single-phase gamma, but oxygen contents of 1000 and 10,000 ppm produce microstructures in which the volume of alpha increases from zero to 77 and 55 per cent, respectively. Nitrogen also has a very noticeable effect.

The gamma phase can be retained completely or partially by quenching alloys containing from 20 to 75 wt.% uranium in water. The retained gamma transforms isothermally to epsilon. The initiation of gamma transformation occurs earlier at compositions above or below the epsilon composition (about 50 wt.% uranium). Except for temperatures just below and above the gamma-epsilon equilibrium temperature ($580^\circ C$), the gamma-to-epsilon transformation is accompanied by a marked hardness increase and an aging peak. In alloys containing less than 20 wt.% uranium, the gamma transforms martensitically on quenching to yield a strained alpha-zirconium lattice.

Zirconium-uranium alloys are of special interest because they can be heat-treated. Alloys

in the 75 to 95 wt.% uranium range are subject to far more microstructural manipulation by heat-treatment than other alloys in the system because of the nature of the phase fields in this composition range. Annealed alloys exhibit mechanical properties intermediate between those of the alpha zirconium and epsilon phases. The mechanical strength of alloys having compositions up to 19 wt.% uranium is sensitive to the size of the alpha-phase particles and the uranium content of the alpha phase. Maximum ductility is obtained by quenching or furnace-cooling alloys with uranium contents close to or beyond the concentration range of the epsilon phase. On the other hand, maximum hardness and minimum ductility are obtained when these alloys are air cooled.

All the alloys in the system can be fabricated by a variety of techniques. The best conditions of fabrication are determined not only by the constitutional diagram and heat-treating characteristics but also by the type of fabrication and by the size and geometry of the piece to be fabricated. Hot-hardness data are useful in understanding and selecting fabrication procedures.

Alloys containing up to 95 wt.% uranium form a tarnish film and exhibit little or no weight change when exposed to 100°C (212°F) water. Such alloys must be quenched rapidly from above 700°C, and the test water must contain sufficient oxygen to maintain a protective oxide film. If the alloys are annealed just below the alpha-beta transformation, only those containing less than 80 wt.% uranium are resistant in boiling water. In general, only alloys containing less than 60 wt.% uranium survive prolonged exposure in 360°C (680°F) water. Zirconium-70 wt.% uranium alloys are fairly resistant to 260°C (500°F) water, whereas alloys containing 90 wt.% uranium can withstand 178°C (352°F) water. Since epsilon is the predominant phase and is located in the middle portion of the diagram (30 to 70 wt.%) and since oxygen affects the stability of the epsilon phase, it follows that oxygen should influence the corrosion rate. In the epsilon range, increased oxygen contents result in increased corrosion rates for a given alloy composition.

The irradiation performance of alloys containing 22 wt.% uranium or less appears to be fairly well defined up to about 425°C (800°F). The volume change per atomic per cent burnup is 7 per cent or less (average about 3 per cent). Above 425°C, the volume increases rapidly with

an increase in temperature. No significant changes in the irradiation behavior of the 22 wt.% uranium alloy can be attributed to variations in the distribution of the phases present.

Only limited irradiation data are available for alloys in the 40 to 60 wt.% uranium range, and this is at burnups of less than 1 at.%. Improved behavior might be expected for the epsilon-phase alloys at elevated temperatures since alloys near the middle of the system exhibit greater strength due to solid-solution strengthening.

Niobium-Uranium Alloys

Battelle²⁵ has stressed niobium-10 wt.% uranium alloys at 1600°F with a load of 40,000 psi; no rupture occurred during a 1000-hr testing period. The average creep rate was 0.0002 per cent per hour (including initial load deformation). Specimens have also been run for 300 hr under a load of 10,000 psi at 2200°F with no perceptible deformation after initial loading.

(R. F. Dickerson)

Plutonium

Data obtained at Hanford²⁷ indicate that plutonium thermally cycled through the low-temperature allotropic transformations exhibited greater physical damage than that reported for any other metal or alloy. Observations showed that increasing the specimen size produced a greater decrease in density as a function of the number of cycles. The degree of void formation was directly proportional to volume change associated with the phase transformations. Other variables such as the length-diameter ratio (constant diameter) had little or no influence on the amount of physical damage. Tensile strength of specimens cycled 10 times between the gamma and alpha phases decreased from 62,800 to 30,000 psi. Yield strength dropped from 38,000 to 26,000 psi.

Plutonium-Zirconium Alloys

The plutonium-zirconium phase diagram has been investigated²⁸ from room temperature to 1300°C. The diagram appears in Fig. 1. Two compounds appear in the system. The compound at about 75 at.% zirconium has a hexagonal P6/*mmm* structure, with $a = 5.055$ Å and $c = 3.123$ Å. The other compound (approximately Pu₂Zr) has an unknown crystal structure and a wide

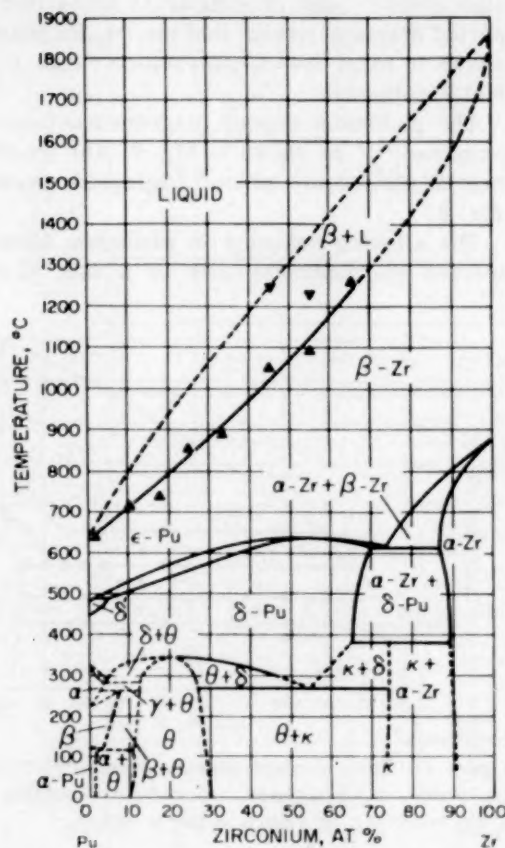


Figure 1—Plutonium-zirconium phase diagram.²⁸ Thermal-analysis results: ▼, liquidus; ▲, solidus.

composition range from about 12 at.% to about 25 at.% zirconium. The delta-phase plutonium structure may readily be retained by quenching samples with compositions between 5 and 70 at.% zirconium, but this phase is not an equilibrium below 270°C. Epsilon-phase plutonium or beta-phase zirconium cannot be retained by quenching at any composition.

Plutonium-Nickel Alloys

The crystal structure of PuNi_4 is reported by Los Alamos²⁹ to be monoclinic (probably space group $C2/m$) with $a \approx 4.86$ Å, $b \approx 8.48$ Å, $c \approx 10.26$ Å, and $\beta \approx 100^\circ$.

Delta-Phase Alloys

A number of binary plutonium alloys³⁰ containing yttrium and rare-earth metals were examined for retention of the delta phase at room temperature. Results to date indicate that cerium, dysprosium, and erbium stabilize the

delta phase; whereas yttrium, lanthanum, praseodymium, neodymium, and gadolinium are nondelta stabilizers. Alloys with 1, 3, and 5 at.% silver additions were similarly studied.²⁹ The delta phase could not be retained in any of the silver-bearing alloys.

Liquid Plutonium Alloys

British³¹ data indicate that a cerium-25 at.% cobalt-14 at.% plutonium alloy, although having a sufficiently low melting point (400 to 420°C) and a desirable plutonium concentration (2 to 3 g of plutonium per cubic centimeter), exhibits some unsatisfactory properties. The solid material has a negative coefficient of expansion, and the molten material is excessively corrosive to container material at about 800°C. Cursory investigations show that low-melting cerium-copper-plutonium and cerium-nickel-plutonium alloys are less corrosive and should be investigated further.

(V. W. Storhok)

Grenoble Plutonium Metallurgy Conference

The Plutonium Metallurgy Conference was held in Grenoble, France, in April 1960. Most of the papers presented at the conference are reviewed here.

Phase Studies of Plutonium-Bearing Materials

Phase diagrams of plutonium with zirconium, neptunium, titanium, lanthanum, and cobalt are presented^{32,1} and are shown in Figs. 2 to 6. The plutonium-zirconium diagram is similar to that reported in the Geneva Conference papers, and the plutonium-cobalt diagram is reported to be in good agreement with the results obtained at Los Alamos. X-ray data for the intermetallic compound phases are reported.

The plutonium-neptunium diagram in the above report is in fair agreement with a partial diagram of the plutonium-rich end of the system shown by Cope et al.^{32,2} However, the beta-plutonium field in this report is shown as exhibiting limited solubility for neptunium. The reasons for the discrepancy are not clear. However, this partial diagram is based on dilatometric and thermal analyses, whereas supplemental X-ray diffraction data were obtained in the previous work.

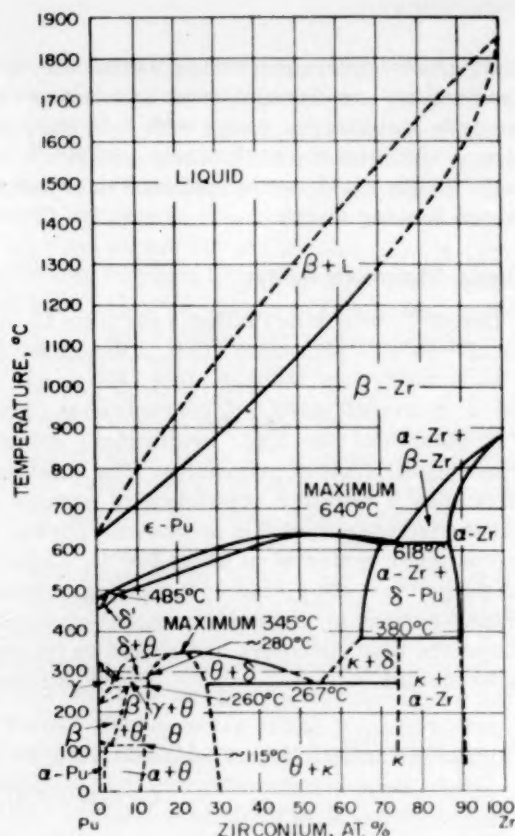


Figure 2—Plutonium-zirconium phase diagram.^{32.1}

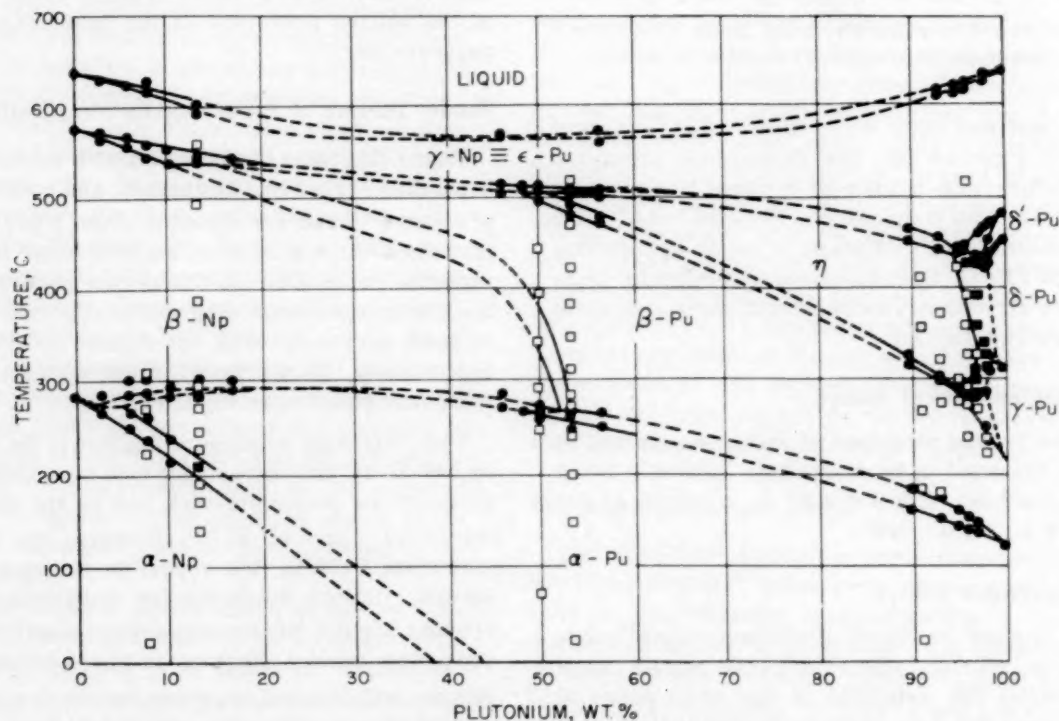


Figure 3—Tentative equilibrium diagram for the neptunium-plutonium system.^{32.1} X-ray identification: □, single phase; ■, two phase. ●, thermal-analysis transitions.

A partial plutonium-ruthenium diagram is also given^{32.2} and is similar to an earlier reported diagram, except that the Pu_{19}Ru phase is shown to exist over a composition range of 3 to 6 at. % ruthenium.

The plutonium-copper diagram has been determined,^{32.3} as shown in Fig. 7. The constitution of plutonium-carbon^{32.4} alloys is shown in Fig. 8.

The alloying behavior of plutonium alloys is treated thermodynamically in a Los Alamos

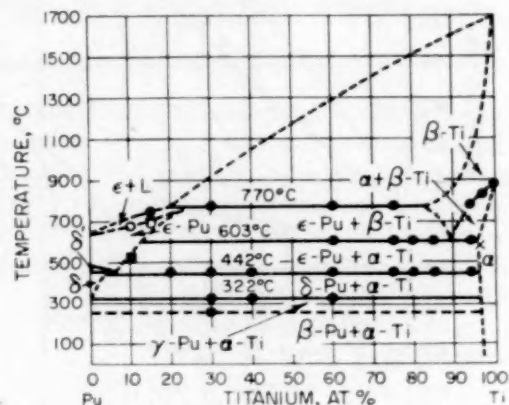


Figure 4—The plutonium-titanium system.^{32.1} ○, single phase. ■, two phase. ●, thermal analysis. ×, X-ray boundary. δ' region is due to Elliott.

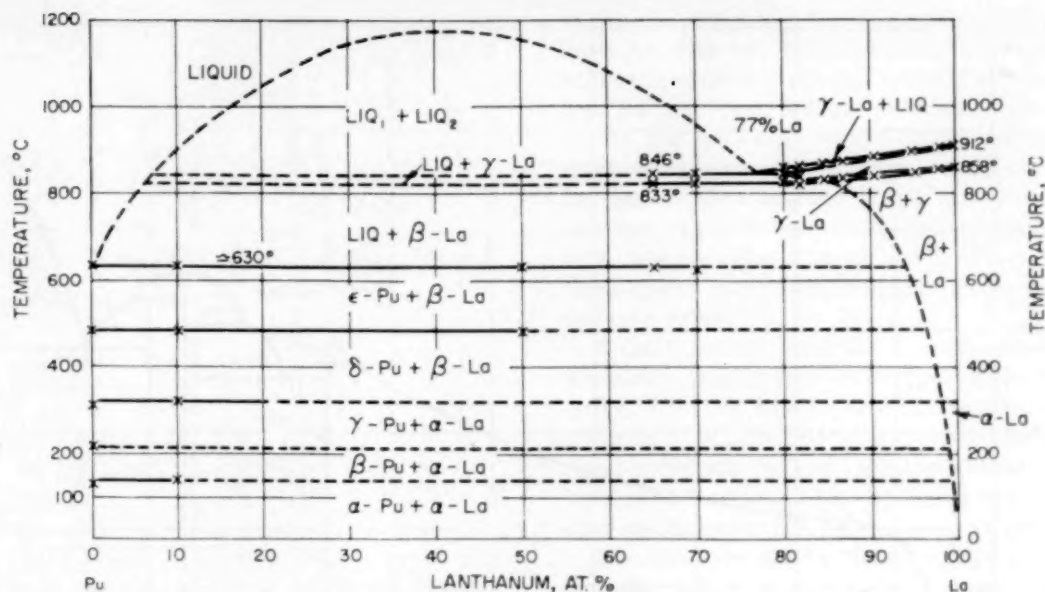


Figure 5—Tentative phase diagram of the plutonium-lanthanum system.^{32,1} ×, thermal-analysis points.

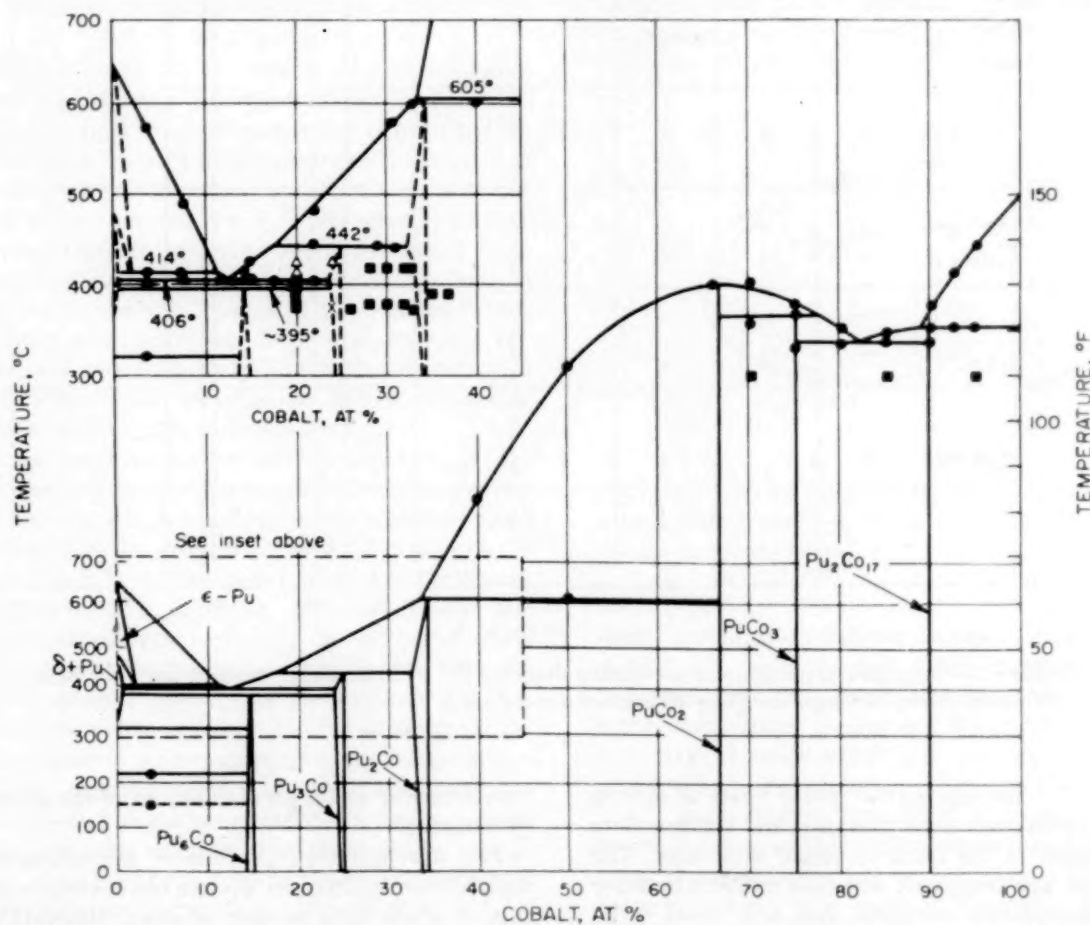


Figure 6—Plutonium-cobalt phase diagram.^{32,1} Metallographic data: ●, single phase; ■, two phase; Δ, partly melted. ×, thermal-analysis point.

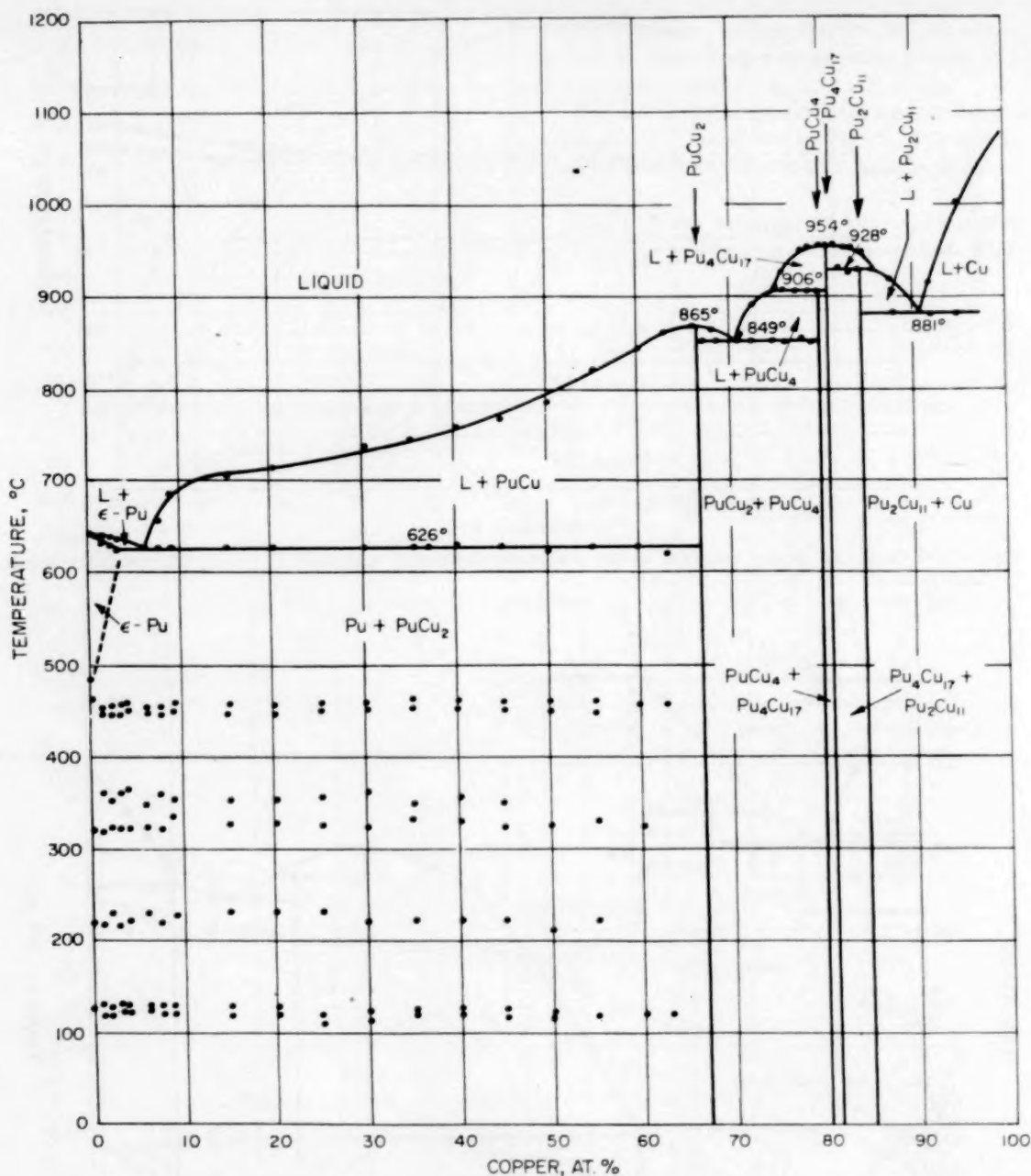
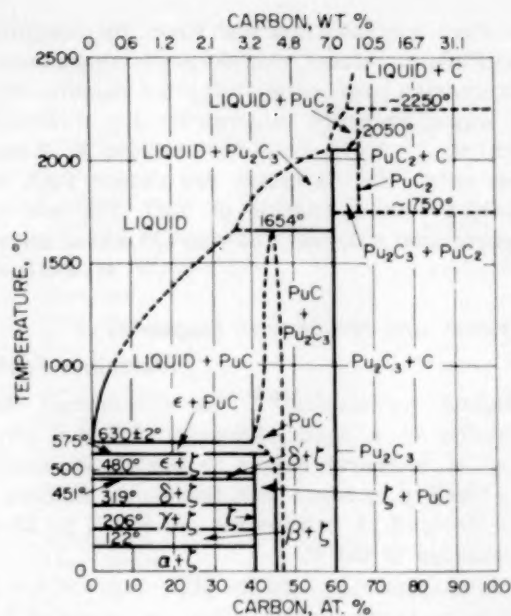


Figure 7—Proposed plutonium-copper binary diagram.^{32.3} ●, differential thermal-analysis data. (All temperature readings are given in degrees centigrade.)

paper.^{32.5} Excess partial molar heats of mixing are calculated from liquidus and solidus data available in the form of phase diagrams. The results are compared with data obtained by other thermodynamic methods and are found to be reasonable. A discussion of the relation between the thermodynamic data and the effects of elec-

tron transfer and electron number of the solute is included.

The uranium-rich portion of the uranium-molybdenum-plutonium system has been the subject of study at a number of sites. Harwell^{32.6} reports on the constitution and fabrication characteristics of these alloys. Isothermal ternary

Figure 8—Plutonium-carbon phase diagram.^{32.4}

sections from 450 to 1000°C are shown along with liquidus and solidus contours at temperatures to 1200°C. A ternary eutectoid, containing from 10 to 14 at.% molybdenum and 17 to 20 at.% plutonium, is reported as occurring at about 525°C. An estimated 8 at.% plutonium is soluble in the intermediate uranium-molybdenum delta phase. Resistance-melting methods, as applied to the ternary alloys, and the results of hot rolling are described. In general, ductility decreases with increasing plutonium content, with the effect most marked in alloys containing more than 15 at.% molybdenum.

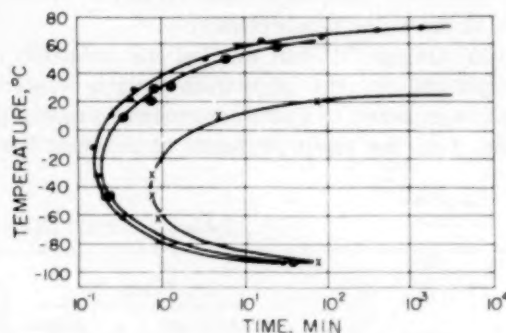
The kinetics of transformation of the gamma uranium phase in uranium-molybdenum-plutonium alloys near the ternary eutectoid composition is the subject of two French papers. In one paper,^{32.7} dilatometric studies of the transformation are reported. Both expansions and contractions are reported, the indication being that transformation is the result of two separate mechanisms. These two mechanisms are described in the second paper^{32.8} as being (1) the formation of a new gamma phase, closely related to the original gamma phase in structure, accompanied by the precipitation of alpha uranium and the zeta uranium-plutonium phase and (2) the ordering of the gamma phase with additional precipitation of the alpha and zeta phases.

The requirements for alloy additions which will permit delta-phase retention in plutonium

alloys are given in a Los Alamos paper.^{32.9} The requirements are a valence of 3 or 4 and size differences from delta plutonium of between -15 and +8 per cent for the trivalent solute elements and between -10 and +3 per cent for the tetravalent Group IVA solute elements. Only cerium, with a valence of 3.6 and a size difference of +3.4 per cent, and divalent zinc, with a size difference of -15.2 per cent, are delta-phase plutonium retainers and exceptions to these rules.

A dilatometric study of the delta-to-gamma plutonium transformation is reported in a French paper.^{32.10} The results indicate that the transformation occurs martensitically. The structural relations between the two phases and the atom shifts required for the transformation are described.

The kinetics of the beta-to-alpha plutonium transformation is described in an Aldermaston paper.^{32.11} The transformation is described as a nucleation and growth process, and the results are summarized in the time-temperature transformation diagram of Fig. 9. The transformation

Figure 9—Beta-to-alpha isothermal transformation kinetics.^{32.11} •, start; ●, 40 per cent transformed; x, 100 per cent transformed.

at any temperature follows an exponential of the form e^{-At^k} , where A and k are constants and t is the time. The formation of voids during alpha-beta phase cycling and the application of pressure to produce 100 per cent dense alpha plutonium are described.

An Aldermaston report^{32.12} also describes the techniques used for the preparation of plutonium for metallographic examination. Cathodic vacuum etching was found to provide a highly satisfactory method for etching samples.

(A. A. Bauer)

Behavior of Plutonium Fuels

The French^{32,13} report a eutectic type reaction that forms at 450°C between stainless steels and alloys of uranium-plutonium-molybdenum. The rate of eutectic formation increases with molybdenum content in these alloys.

Sackman^{32,14} reports that plutonium metal oxidizes interfacially. Initial oxidation of alpha plutonium forms a black oxide that is progressively replaced by a yellow dioxide. Complete specimen oxidation gives a weight gain greater than would be the case if the samples were completely converted to PuO₂. A delta-stabilized plutonium alloy (3.5 per cent aluminum) has greater corrosion resistance. It appears that the corrosion mechanism of alpha plutonium is an intergranular one, whereas a uniform attack occurs on delta plutonium.

A paper by the British^{32,15} at Harwell reports the thermal-expansion coefficients of PuO₂ in the temperature range of room temperature through 1000°C obtained using an argon-atmosphere dilatometer. The value given for the thermal expansion of plutonium oxide at 1000°C was 1.063 per cent, as compared with 1.010 per cent for UO₂ at the same temperature.

Los Alamos^{32,16} has studied the volatility of compounds in the plutonium-oxygen system. PuO₂ was shown to be the congruently vaporizing solid, i.e., the vapor pressure at constant tem-

perature was invariant with time. By assuming that PuO₂ molecules were the predominant gaseous species over solid PuO₂, the equation for the vapor pressure is given by: $\log P \text{ (atm)} = -29,240/T + 8.072$, from 2000 to 2400°K. It was also noted that tantalum can reduce PuO₂ to Pu₂O₃ by the formation of TaO. The heat of vaporization was reported to be 133.8 kcal/mole.

(W. M. Pardue)

Physical and Mechanical Properties of

Plutonium Fuels

Mound Laboratory^{32,17} has determined the viscosity of a liquid plutonium-9.5 at.% iron alloy at temperatures up to 808°C. Viscosity was found to increase with decreasing temperature from 6.14 centipoises at 808°C to 22.4 centipoises at 433°C.

Cubic-expansion data for plutonium and for a plutonium-iron eutectic alloy, as reported by Los Alamos,^{32,18} are shown in Tables I-4 to I-6. The relatively large data differences noted between the two plutonium-iron eutectic alloys suggest that the expansion properties in the solid state are sensitive to impurity content, fabrication history, and/or plutonium-iron ratio.

Thermal expansion of delta-phase plutonium solid-solution alloys containing zinc, aluminum, cerium, and zirconium has been investigated.^{32,19} Results indicate that initial, small amounts of

Table I-4 CUBIC EXPANSION OF PLUTONIUM METAL^{32,18}

| Phases | Temp., °C | | Density, g/cm ³ | Coefficient of cubic expansion from low to high temperature in phase ($\times 10^{-6}$) | Expansion, vol. % |
|---------------------|------------------|-----------------|--|---|-------------------|
| | At phase changes | At data points | | | |
| Alpha | | 24.0 116.75 | 19.64 19.3 ₃₅ | 170 | |
| Alpha-beta | 122 ± 2 | | | | 8.6 ₅ |
| Beta | | 132.75 192.0 | 17.7 ₄₉ 17.5 ₉₂ | 150 | |
| Beta-gamma | 206 ± 3 | | | | 2.4 ₈ |
| Gamma | | 217.0 300.5 | 17.0 ₉₇ 16.9 ₄₀ | 111 | |
| Gamma-delta | 319 ± 5 | | | | 6.8 ₄ |
| Delta | | 327.5 435.0 | 15.8 ₂₇ 15.8 ₈₆ | -34 | |
| Delta-delta prime | 451 ± 4 | | | | Delta to epsilon |
| Delta prime | | 462.75 | 15.9 ₂₅ | | -3.46 |
| Delta prime-epsilon | 480 ± 7 | | | | |
| Epsilon | | 492.0 626.5 | 16.4 ₄₄ 16.2 ₇₇ | 85 | |
| Epsilon-liquid | 639.5 ± 2 | | | | -0.8 ₂ |
| Liquid | | 648.0 707.8 | 16.3 ₇₃ 16.2 ₂₁ | 157 | |

Table I-5 CUBIC EXPANSION OF A PLUTONIUM-2.36 WT.% IRON ALLOY^{32,18}

| Phase (+ Pu ₄ Fe) | Temp., °C | | Density, g/cm ³ | Coefficient of cubic expansion from low to high temperature in phase ($\times 10^{-6}$) | Expansion, vol.% |
|---------------------------------|---------------------|-------------------|-------------------------------|--|---------------------|
| | At phase changes | At data points | | | |
| Alpha | | 25 | 16.857 | | |
| Alpha-beta | 122 \pm 2 | | | | |
| Beta | | 138 | 16.50 | 133. ₈ | |
| | | 150 | 16.47 | | |
| | | 193 | 16.38 | | |
| Beta-gamma | 206 \pm 3 | | | | 0.3 ₁ |
| Gamma | | 225 | 16.26 | 137. ₅ | |
| | | 308 | 16.08 | | |
| Gamma-delta | 319 \pm 5 | | | | 0.9 ₃ |
| Delta | | 328 | 15.90 | 65. ₉ | |
| | | 412 | 15.81 | | |
| Delta-liquid | 411 \pm ? | | | | -1.9 ₆ |
| Liquid (only) | | 470 | 16.01 | 124. ₄ | |
| | | 535 | 15.88 | | |

Table I-6 CUBIC EXPANSION OF A PLUTONIUM-2.25 WT.% IRON ALLOY^{32,18}

| Phase (+ Pu ₄ Fe) | Temp., °C | | Density, g/cm ³ | Coefficient of cubic expansion from low to high temperature in phase ($\times 10^{-6}$) | Expansion, vol.% |
|---------------------------------|---------------------|-------------------|-------------------------------|--|---------------------|
| | At phase changes | At data points | | | |
| Alpha | | 23.5 | 16.770 | 112. ₃ | |
| | | 109.75 | 16.61 | | |
| Alpha-beta | 122 \pm 2 | | | | 0.4 ₁ |
| Beta | | 136.5 | 16.49 | 89. ₂ | |
| | | 197.25 | 16.40 | | |
| Beta-gamma | 206 \pm 3 | | | | 0.2 ₄ |
| Gamma | | 221.5 | 16.32 | 76. ₃ | |
| | | 296 | 16.23 | | |
| Gamma-delta | 319 \pm 5 | | | | 1.4 ₈ |
| Delta | | 329 | 15.96 | 138. ₄ | |
| | | 400 | 15.80 | | |
| Delta-liquid | 411 \pm ? | | | | -2.1 ₇ |
| Liquid (only) | | 460 | 16.04 | 113. ₃ | |
| | | 621 | 15.76 | | |
| | | 664 | 15.68 | | |

Table I-7 ELASTIC CONSTANTS FOR PLUTONIUM METAL^{32,21}

| Remarks | Density, g/cm ³ | Length, cm | Diameter, cm | Young's modulus (E), 10 ¹¹ dynes/cm ² | Modulus of rigidity (G), 10 ¹¹ dynes/cm ² | Poisson's ratio |
|--------------|-------------------------------|---------------|-----------------|---|---|-------------------|
| As cast | 19.50 \pm 0.02 | 2.541 | 0.30 | 9.76 \pm 0.01 | 4.15 \pm 0.005 | 0.175 \pm 0.005 |
| As cast | 19.50 | 2.542 | 0.30 | 9.77 | 4.15 | 0.177 |
| As cast | 19.48 | 2.250 | 0.30 | 9.84 | 4.17 | 0.181 |
| High density | 19.71 | 2.021 | 0.31 | 10.04 | 4.23 | 0.186 |
| High density | 19.74 | 1.502 | 0.30 | 9.76 | | |

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Thermal expansion of delta-phase plutonium solid-solution alloys containing zinc, aluminum, cerium, and zirconium has been investigated.^{32,19} Results indicate that initial, small amounts of

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|---------------------|------------------|----------------|----------------------------|---|-------------------|
| | At phase changes | At data points | | | |
| Alpha | | 24.0 | 19.64 | 170 | |
| | | 116.75 | 19.3 ₃₅ | | |
| Alpha-beta | 122 ± 2 | | | | 8.6 ₅ |
| Beta | | 132.75 | 17.7 ₄₈ | 150 | |
| | | 192.0 | 17.5 ₉₂ | | |
| Beta-gamma | 206 ± 3 | | | | 2.4 ₈ |
| Gamma | | 217.0 | 17.0 ₉₇ | 111 | |
| | | 300.5 | 16.9 ₄₀ | | |
| Gamma-delta | 319 ± 5 | | | | 6.8 ₄ |
| Delta | | 327.5 | 15.8 ₂₇ | -34 | |
| | | 435.0 | 15.8 ₈₅ | | |
| Delta-delta prime | 451 ± 4 | | | | Delta to epsilon |
| Delta prime | | 462.75 | 15.9 ₂₅ | | -3.46 |
| Delta prime-epsilon | 480 ± 7 | | | | |
| Epsilon | | 492.0 | 16.4 ₈₄ | 85 | |
| | | 626.5 | 16.2 ₇₇ | | |
| Epsilon-liquid | 639.5 ± 2 | | | | -0.8 ₂ |
| Liquid | | 648.0 | 16.3 ₇₅ | 157 | |
| | | 707.8 | 16.2 ₂₁ | | |

Table I-5 CUBIC EXPANSION OF A PLUTONIUM-2.36 WT.% IRON ALLOY^{22,18}

| Phase (+ Pu ₂ Fe) | Temp., °C | | Density, g/cm ³ | Coefficient of cubic expansion from low to high temperature in phase (× 10 ⁻⁴) | Expansion, vol. % |
|---------------------------------|---------------------|-------------------|-------------------------------|---|----------------------|
| | At phase changes | At data points | | | |
| Alpha | | 25 | 16.857 | | |
| Alpha-beta | 122 ± 2 | | | | |
| Beta | | 138 | 16.50 | 133. ₃ | |
| | | 150 | 16.47 | | |
| | | 193 | 16.38 | | |
| Beta-gamma | 206 ± 3 | | | | 0.3 ₁ |
| Gamma | | 225 | 16.26 | 137. ₅ | |
| | | 308 | 16.08 | | |
| Gamma-delta | 319 ± 5 | | | | 0.9 ₃ |
| Delta | | 328 | 15.90 | 65. ₉ | |
| | | 412 | 15.81 | | |
| Delta-liquid | 411 ± 7 | | | | -1.9 ₈ |
| Liquid (only) | | 470 | 16.01 | 124. ₄ | |
| | | 535 | 15.88 | | |

Table I-6 CUBIC EXPANSION OF A PLUTONIUM-2.25 WT.% IRON ALLOY^{22,18}

| Phase (+ Pu ₆ Fe) | Temp., °C | | Density, g/cm ³ | Coefficient of cubic expansion from low to high temperature in phase (×10 ⁻⁶) | Expansion, vol. % |
|---------------------------------|---------------------|-------------------|-------------------------------|--|----------------------|
| | At phase changes | At data points | | | |
| Alpha | | 23.5 | 16.770 | 112. ₉ | |
| | | 109.75 | 16.61 | | |
| Alpha-beta | 122 ± 2 | | | | 0.4 ₇ |
| Beta | | 136.5 | 16.49 | 89. ₂ | |
| | | 197.25 | 16.40 | | |
| Beta-gamma | 206 ± 3 | | | | 0.2 ₈ |
| Gamma | | 221.5 | 16.32 | 76. ₃ | |
| | | 296 | 16.23 | | |
| Gamma-delta | 319 ± 5 | | | | 1.4 ₉ |
| Delta | | 329 | 15.96 | 138. ₄ | |
| | | 400 | 15.80 | | |
| Delta-liquid | 411 ± 7 | | | | -2.1 ₇ |
| Liquid (only) | | 460 | 16.04 | 113. ₃ | |
| | | 621 | 15.76 | | |
| | | 664 | 15.68 | | |

Table I-7 ELASTIC CONSTANTS FOR PLUTONIUM METAL^{22,21}

| Remarks | Density, g/cm ³ | Length, cm | Diameter, cm | Young's modulus (E), 10 ¹¹ dynes/cm ² | Modulus of rigidity (G), 10 ¹¹ dynes/cm ² | Poisson's ratio |
|--------------|-------------------------------|---------------|-----------------|---|---|-------------------|
| As cast | 19.50 \pm 0.02 | 2.541 | 0.30 | 9.76 \pm 0.01 | 4.15 \pm 0.005 | 0.175 \pm 0.005 |
| As cast | 19.50 | 2.542 | 0.30 | 9.77 | 4.15 | 0.177 |
| As cast | 19.48 | 2.250 | 0.30 | 9.84 | 4.17 | 0.181 |
| High density | 19.71 | 2.021 | 0.31 | 10.04 | 4.23 | 0.186 |
| High density | 19.74 | 1.502 | 0.30 | 9.76 | | |

alloy additions to plutonium result in an increase in the rate of negative expansion of the delta phase. Further additions reverse this effect, the expansion becoming zero and then positive with increasing alloy content.

A British^{32,20} paper presented the results of a number of property studies of possible fuel alloys consisting of plutonium-iron, zirconium, thorium, uranium, and uranium-molybdenum. Oxidation behavior of the alloys was determined in air up to 500°C and in carbon dioxide up to 800°C. Compatibility studies of ternary uranium-molybdenum-plutonium alloys with stainless steel revealed a marked rate of attack between 600 and 650°C. This may have been caused by the formation of a low-melting plutonium-iron eutectic. Also, room-temperature hardness was obtained on all alloys, and hot-hardness figures are presented for the uranium-molybdenum-plutonium alloys.

Young's modulus and the rigidity modulus for plutonium metal are shown in Table I-7.^{32,21}

Chalk River^{32,22} reports data obtained during the initial assessment of aluminum-plutonium alloys sheathed with Zircaloy-2. Nominal compositions studied were aluminum-5, 10, 15, and 20 wt.% plutonium. In the temperature range of interest (300 to 400°C), there was an insignificant reaction between the core and the Zircaloy sheath. Corrosive attack on base alloys in 340°C deionized water varied from approximately 0.008 cm/hr for the 5 wt.% plutonium alloy to 1.5×10^{-5} cm/hr for the 20 wt.% plutonium alloy. Alloys sheathed in Zircaloy-2 cans underwent less attack. Sheathed alloys were irradiated for four months under power-reactor conditions at a temperature of approximately 400°C and a total atom burnup of 0.2 per cent. The density of the core decreased by about 2 per cent, whereas hardness increased.

Hanford^{32,23} has studied the effect of temperature and testing speed on the tension and compression properties of unalloyed plutonium in the alpha, beta, gamma, and delta phases. In general, the effect of increasing temperature was to decrease strength and to increase ductility. Varying effects were produced by changing the testing speed. Some creep, tension impact, and torsion data for alpha-phase plutonium are reported. In addition, extrusion constants and pressures for the beta, gamma, and delta phases were obtained. (V. W. Storhok)

Melting and Fabrication of

Metallic Plutonium Fuel Materials

Consideration is given by Vallecitos Atomic Laboratory^{32,24} to the methods by which plutonium is purified at each chemical process step in converting plutonium nitrate to metal and the subsequent melting and casting of the metal into an appropriate shape for shop operations. The degree of purification that may be obtained in each chemical process step is discussed in a qualitative way on the basis of laboratory investigations or plant operating experience.

In the metallurgical phase of the process, melting of the metal and casting to shape, the removal of impurities by the oxide-dross technique, and the removal of impurities by volatilization is considered. The amount of purification that may be achieved by the oxide-dross technique is discussed on the basis of operating experience with both plutonium and uranium. Calculations are made to determine which impurities can be removed from molten plutonium by volatilization. For this purpose, the Langmuir equation for the rate of evaporation from a surface into a vacuum is utilized on the basis that it applies to ideal solutions as well as to pure metals.

Twenty-five disks of delta-stabilized plutonium with close-dimensional tolerances were required for criticality measurements for the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE).^{32,25} These disks were 0.125 in. thick and 5.937 in. in diameter. They were to be handled and tested in equipment and laboratories used for purposes other than plutonium work; therefore, it was necessary that they be in tight alpha-containing cans.

Two methods were used for making these disks: (1) blanking from sheet stock made by extrusion and casting and (2) direct machining. In the blanking method, a cast feed ring was rough machined to weight and then extruded into a tube of the proper wall thickness in a specially designed direct-extrusion die assembly. This tube was split and flattened. Disks were cut from the roughly flattened sheet with a blanking die in a slow-moving hydraulic press. The disks were then flattened and pressed to exact size in a coining die. In the machining method, an oversized disk was cast in an alumina-coated graphite crucible in a vacuum furnace. This casting was machined to dimensions in a lathe.

The disks were canned in 0.005-in.-thick nickel cans which had been formed by blanking and drawing. The disks were carefully assembled in the cans, and the edges were sealed with a lead-tin solder.

The preparation of high-purity plutonium has been investigated at Argonne by fused-salt electrolysis with plutonium anodes containing 1000 to 4000 ppm of impurities.^{32,26} Electrolyses have been carried out in Hastelloy C and Vycor cells and with molybdenum and tantalum cathodes. Good buttons have been obtained by consolidation of the deposits by melting under molten LiCl-KCl or NaCl-KCl. Over-all recoveries greater than 60 per cent have been achieved.

High-purity plutonium prepared elsewhere by bomb reduction contains at least 300 ppm of impurities, whereas electrolytic metal with 65 ppm of analyzed impurities has been prepared with Vycor cells and tantalum cathodes.

Major remaining impurities are hydrogen, carbon, oxygen, and uranium. The effects of temperature, electrolysis current, meltdown salt composition, and salt washing of deposits before meltdown on the purity of the final product have also been studied. Vacuum melting of plutonium in MgO crucibles has caused a further reduction in the hydrogen contamination but has not caused an observable magnesium contamination.

Metallographic examination of impurities in metal produced by electrolysis seems to be in agreement with the chemical analyses.

A brief look was also taken at the possibilities for producing massive plutonium of improved purity by directional solidification in vacuum. The hydrogen content of plutonium treated in this manner has been reduced to 2 to 3 ppm without further magnesium or oxygen contamination. Vertical variation of the concentrations of chromium, iron, and manganese has been noted with each of these elements more concentrated near the top of the directionally solidified ingot. The possibility exists, therefore, of reducing these impurities by this technique. Carbon was apparently unaffected.

An English paper^{32,27} describes some of the initial experiments on the rolling of plutonium and the tensile testing of the sheet produced.

Small ingots measuring 2 by 2 by 0.25 in. were prepared by melting in a tantalum crucible and casting under an argon atmosphere into alumina-coated graphite molds heated to 300°C.

Because of the brittle nature of plutonium in the alpha range, initial experiments were directed toward rolling preheated ingots with heated rolls in the upper beta range. Both ingots and rolls were heated to 175°C.

The two rolling schedules chosen were apparently equally successful. In the first schedule the ingots were rolled from 0.225 to 0.048 in., a total of 78 per cent, in 15 passes with the heaviest single pass being 12 per cent. In the second schedule the ingots were given a total of 75 per cent reduction in five passes, which involved individual pass reductions up to 34 per cent. With each schedule a slight amount of edge cracking occurred.

A small specimen cut from sheet produced from the first schedule was subsequently reheated to 175°C and rolled satisfactorily in five passes to 0.16 in.

Considerable distortion and surface marking resulted in all sheet material when it was cooled down through the beta-alpha transformation temperature. Distortion could be eliminated by loosely clamping the material between two warmed steel plates and allowing it to cool. Warm rolling in the high alpha range (105°C) to a reduction of between 7 and 16 per cent resulted in the elimination of both distortion and surface marking, but at the expense of cracking if the sheet entering the rolls were not flat.

Tensile tests were carried out over a temperature range of 25 to 400°C on specimens prepared from sheet rolled in the beta phase and cooled to room temperature. On the basis of these tensile tests, it appears that the metal is extremely ductile at high temperatures in the beta range with elongation in the region of 350 per cent. The general form of the delta-phase load extension curves is indicative of hot working and implies that deformation is accompanied by recrystallization.

The properties of the metal in the beta phase are temperature dependent, and the ultimate tensile strength increases markedly as the temperature is lowered; following a beta-phase anneal at 175°C, the elongation values decrease with decreasing temperature, particularly below 1300°C, but it is anticipated that the elongation values obtained may be highly sensitive to strain rate.

Aluminum-clad, aluminum-10 wt.% plutonium alloy fuel elements have been produced by the French using the picture-frame roll-bonding process.^{32,28}

The cores are produced by casting and machining, and, after ultrasonic cleaning, they are removed from the glove boxes and assembled with the picture frames and cover plates and welded. The outgassed assemblies are subjected to a hot- and cold-rolling schedule, radiographed to determine core location, rolled into a cylinder, and attached to an aluminum positioner.

Tests are in progress to utilize an aluminum-13 wt.% silicon alloy interface layer to improve the bonding between the core edge and picture frame.
(R. J. Carlson)

Fabrication of Nonmetallic Plutonium Fuels

Several attempts^{32,29,32,30} have been made to investigate the sintering behavior of UO_2 - PuO_2 mixtures. Sinterability of physical mixtures of the oxides decreases with increasing PuO_2 content to 10 wt.%, with the most drastic effect occurring below 1 wt.% PuO_2 . Sinterability increases with increasing PuO_2 content when the PuO_2 is added to UO_2 in the form of a solid solution ($\text{U}:\text{Pu} = 5:1$). The solid solution was formed by simultaneous coprecipitation of $\text{Pu}(\text{OH})_4$ and $(\text{NH}_4)_2\text{U}_2\text{O}_7$ from hot ammonium hydroxide followed by calcining. For UO_2 -rich compositions, the solid-solution mixture showed a great increase in density when processed by a sintering operation that was followed by grinding and resintering. Conversely, the density decreased in the case of PuO_2 -rich compositions.

PuO_2 is a very stable oxide with a free energy of formation reported to be -240 kcal/mole at room temperature. No reduction is reported when PuO_2 is sintered in dry hydrogen.

(W. M. Pardue)

Thorium

The effect of impurities and alloying elements upon the miscibility gap at 915°C between body-centered cubic thorium containing about 40 wt.% zirconium and body-centered cubic thorium containing about 25 wt.% zirconium has been studied by Murray.³³ The miscibility gap could not be detected in alloys prepared from iodide thorium. The gap was observed, however, in alloys prepared from thorium containing 3000 to 4000 ppm oxygen and 400 to 700 ppm carbon. Alloy additions of indium, uranium, and titanium caused the miscibility gap to expand in terms of composition and in terms of the maximum tempera-

ture at which the gap is observed. Alloy additions of cerium and hafnium apparently reduced the stability of the body-centered cubic thorium solution at 25 wt.% zirconium such that the monotectoid reaction at 915°C disappeared, and the extent of the high-temperature body-centered cubic solution was restricted.

The lattice spacing of iodide thorium has been determined by Evans and Raynor³⁴ to be 5.0741 ± 0.0002 kx (5.0843 Å). Larger lattice spacings were found for thorium contaminated with nitrogen.

Ames Laboratory³⁵ has determined the electrical resistivity and specific heat of crystal-bar thorium, as follows:

| Temp., °C | Resistivity, $\mu\text{ohm-cm}$ | Specific heat, cal/(mole)(°C) |
|-----------|---------------------------------|-------------------------------|
| 25 | 16.95 | 6.56 |
| 100 | 21.45 | 6.75 |
| 200 | 27.35 | 6.99 |
| 300 | 33.20 | 7.24 |
| 400 | 38.85 | 7.49 |
| 500 | 44.20 | 7.73 |
| 600 | 49.20 | 7.99 |
| 700 | 53.80 | 8.28 |
| 800 | 58.05 | 8.62 |
| 900 | 62.00 | 9.02 |
| 1000 | 65.75 | 9.54 |

The effect of alloying and the effects of fabrication procedures and heat-treatment upon the mechanical and physical properties of thorium-uranium alloys are being investigated at Battelle.^{25,36} It has been found that 10 wt.% additions of zirconium to a thorium-10 wt.% uranium alloy produce a greater strengthening effect in short-time tensile tests at 600°C than 2 wt.% additions of niobium or molybdenum. This effect is dramatically reversed in creep tests at 600°C. This reversal is attributed to the properties of the grain-boundary network of uranium found in these alloys and to the relative effects of additions of niobium, molybdenum, and zirconium upon the strength of this network. A quaternary alloy containing both zirconium and niobium had an ultimate strength of 25,000 psi at 600°C, but creep data are not yet available. It was found that relatively large additions of zirconium only reduce the corrosion rate of thorium-uranium alloys in water at 300°C by a factor of 2. No hope is offered for further major improvements in the corrosion behavior of these alloys. Alloys of thorium and uranium are stated to show finer, more random dispersion of the uranium particles in the thorium when the alloys are prepared

by arc melting than when prepared by induction melting.

Hanford³⁷ reports the following properties for unirradiated thorium and for thorium irradiated to 0.8 at.% burnup in the MTR. The conditions of irradiation are not stated, but the specimen was probably fairly cold. The unirradiated thorium is apparently annealed, bomb-reduced thorium.

| Material history | Yield strength (0.2% offset), 1000 psi | Tensile strength, 1000 psi | Elongation, % | Hardness, R _A |
|------------------|--|-------------------------------|------------------|-----------------------------|
| Unirradiated | 18.3 | 26.7 | 38 | 9 |
| Irradiated | 69.1 | 75.2 | 2 | 53 |

(W. Chubb)

Dispersion Fuel Materials

An investigation by NASA³⁸ of the compatibility of UO₂ with 30 high-melting metals, oxides, carbides, borides, and nitrides was undertaken to indicate combinations that may be suitable for a dispersion type fuel element for a nuclear reactor. Tests were conducted *in vacuo* or in a dried argon atmosphere for 10 min in the 3500 to 5000°F temperature range.

Hafnium carbide, tantalum nitride, and the metals tungsten and tantalum were found to be compatible with UO₂ to its melting point. In addition, ditungsten boride, molybdenum carbide, and the metals columbium and molybdenum were found to be compatible to their melting points.

Battelle³⁹ reports the thermal-conductivity values of 80 vol.% UO₂ cermets. The thermal-conductivity specimens were fabricated by the pressure bonding of green-pressed cores of UO₂-metal mixes for 3 hr at 2300°F under a helium-gas pressure of 10,000 psi. Table I-8 shows the measured mean thermal-conductivity values and gives descriptions of the cermets.

(D. L. Keller)

Refractory Fuel and Fertile Materials

Properties and Behavior of

Uranium Oxide Fuels

Several fine reviews of the uranium-oxygen system and UO₂ fuel performance have been prepared by British and American workers.⁴⁰⁻⁴³

Table I-8 MEAN THERMAL-CONDUCTIVITY VALUES OF UO₂ CERMETS³⁹

| Temp., °C | Thermal conductivity, watts/(cm)(°C) | | |
|-----------|--------------------------------------|------------------|------------------|
| | Specimen TC-102* | Specimen TC-103† | Specimen TC-104‡ |
| 100 | 0.075 | 0.085 | 0.171 |
| 200 | 0.071 | 0.076 | 0.162 |
| 300 | 0.070 | 0.072 | 0.156 |
| 400 | 0.073 | 0.073 | 0.152 |
| 500 | 0.078 | 0.079 | 0.151 |
| 600 | 0.087 | 0.088 | 0.155 |
| 700 | 0.098 | 0.100 | 0.163 |
| 800 | 0.112 | 0.113 | 0.175 |
| 900 | 0.123 | 0.127 | 0.190 |

*Composition: 80 vol.% - 100 + 140-mesh high-fired UO₂-type 302B stainless steel; density is 98.4 per cent of theoretical.

†Composition: 80 vol.% - 100 + 140-mesh depleted spherical UO₂-type 302B stainless steel; density is 97.2 per cent of theoretical.

‡Composition: 80 vol.% - 100 + 140-mesh depleted spherical UO₂-chromium; density is 97.1 per cent of theoretical.

Koenig⁴³ has summarized many physical properties of UO₂ in convenient tabular form.

In a Bettis study⁴⁴ the effects of comminution method and oxidizing and annealing conditions on the density, surface area, and particle-size distribution of UO₂ powders have been determined. Electron microscopy and metallographic techniques used in structural studies of UO₂ are reviewed by Bettis⁴⁵ and Hanford workers.⁴⁶ A differential calorimetry study⁴⁷ of stored energy in irradiated uranium oxides is reported by Childs and McGurn. Of the three irradiated materials, UO₂, U₄O₉, and UO_{2.08} (UO₂ + U₄O₉), only UO_{2.08} shows a significant postirradiation release of stored energy. This material showed a release of 2.4 cal/g at 340°C. Oxygen atoms displaced into the UO₂ phase are believed to reform U₄O₉ at this temperature and liberate the stored energy. The specimens were irradiated up to 6×10^{19} nvt at temperatures less than 100°C. Compatibility of UO₂ pellets in contact with M-257 and 1100 aluminum has been studied by Langrod.⁴⁸ No reaction was observed after 30 days at 1100°F.

Oak Ridge⁴⁹ has reported work of particular interest to the problem of solid-fission-product mobility in high-temperature UO₂-fueled reactors. Postirradiation release of the following species from UO₂ pellets of 85 per cent of the theoretical density has been observed in a helium atmosphere.

| Temp., °F | Release of indicated species, % | | | | | |
|--------------|---------------------------------|------------------|------------------|-------------------|------------------|-------------------|
| | Nb ⁹⁵ | Zr ⁹⁵ | I ¹³¹ | Cs ¹³⁷ | Sr ⁸⁹ | Ru ¹⁰³ |
| 2100 | 0.16 | 0.05 | 12.4 | 11.4 | 0.02 | 0.007 |
| 2500 | 0.005 | 0.007 | 7.4 | 5.5 | 0.01 | 0.001 |

Swedish workers⁵⁰ have demonstrated the strong effect of the O/U ratio on the postirradiation release of Xe¹³³ from very fine neutron-activated UO₂ powders. Various starting materials were oxidized and reduced to give desired O/U ratios. The O/U ratios were measured before and after the postirradiation heat-treatment, and no changes were observed. The diffusion constant follows the familiar relation:

$$D = D_0 \exp(-Q/RT)$$

The diffusion constant and D_0 are plotted in Figs. 10 and 11, respectively. At a given temperature the diffusion constant increases rapidly with the O/U ratio. Although no data are presented, it is stated in this publication that measurements of simultaneously released I¹³³ and Xe¹³³ give diffusion constants and activation energies similar to those for Xe¹³³ alone. This Xe¹³³ precursor would appear to have a large influence on the interpretation of the xenon-release data from these experiments. It is hoped that the iodine results will be published in the near future. With the emphasis on high-temperature fuels, there is a real need for more data on the release of halogens and other nonrare-gas fission products.

Experiments on the diffusion of Xe¹³³ in Core 2 type UO₂ fuel platelets have been completed at Bettis.⁵¹ The equation for Xe¹³³ diffusion is given as $D = 6.6 \times 10^{-6} \exp(-71,700/RT)$. Studies are also being conducted on the diffusion of Kr⁸⁵ in stoichiometric and nonstoichiometric UO₂. Diffusion anneals at 1000°C on samples from irradiation tests showed that the apparent diffusion coefficient for Kr⁸⁵ increases considerably with burnup.

Measurements of the self-diffusion coefficient of uranium in UO₂ by the method of surface-activity decrease have been completed at Bettis.⁵¹ The diffusion coefficient equation can be written as $D = 4.3 \times 10^{-4} \exp(-88,000/RT)$.

In a continuing study of oxide additions to uranium oxide and the effects of these additions on the oxidation and volatilization behavior, Battelle⁵² reports air-oxidation results with La₂O₃-Y₂O₃-CaO additives (Tables I-9 and I-10). Sintered compacts having the higher CaO con-

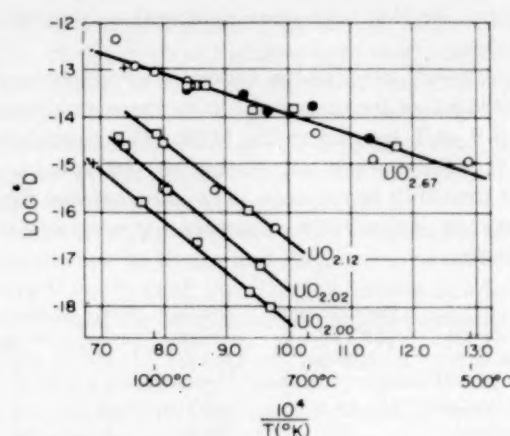


Figure 10—Diffusion constant of Xe¹³³ versus the reciprocal of the absolute temperature.⁵⁰ Starting materials: ○, UO₂, 0.63-μ mean particle size, obtained from AB Atomenergi, Stockholm, and oxidized to UO_{2.12} or U₃O₈; ●, U₃O₈, source unknown, 0.65-μ mean particle size; □, UO_{2.00}, 0.14-μ mean particle size, obtained from AERE, Harwell, reduced to UO_{2.02} or UO_{2.00} or oxidized to UO_{2.12} or U₃O₈; +, UO_{2.18}, 0.05-μ mean particle size, obtained from AERE, Harwell, reduced or oxidized to desired composition. (Reprinted by permission from *Zeitschrift für Naturforschung*.)

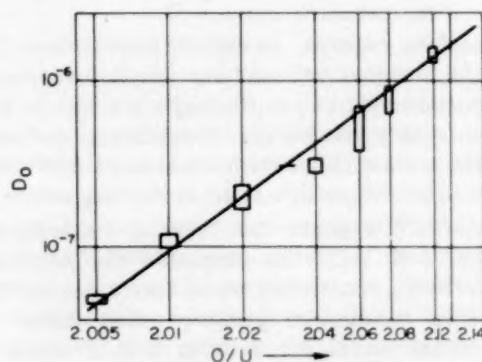


Figure 11—Diffusion constant of Xe¹³³ versus the O/U ratio.⁵⁰ (Reprinted by permission from *Zeitschrift für Naturforschung*.)

tents show better resistance to oxidation. The ultrahigh-pressure study of the uranium-oxygen system is being extended to the UO₂-Al₂O₃ and/or MgO systems.²⁵

Sylvania-Corning workers have summarized⁵³ their work on improving the thermal conductivity of UO₂ with ceramic additives; only solid solutions were studied. Thermal conductivities of sintered UO₂ containing Y₂O₃ and Nb₂O₅ are reported in Fig. 12. The Nb₂O₅ compacts were

Table I-9 SUMMARY⁵² OF WEIGHT CHANGES IN SINTERED TERNARY OXIDE COMPACTS AFTER AIR FIRING AT 1750°C

| Identification | Intended composition, mole % | | | | Weight loss* after heating at 1750°C in dry flowing tank air for indicated time, % | | |
|----------------|------------------------------|--------------------------------|-------------------------------|------|--|-------|-------|
| | UO ₂ | La ₂ O ₃ | Y ₂ O ₃ | CaO | 1 hr | 5 hr | 20 hr |
| 14795-67 | 50.0 | 25.0 | | 25.0 | +1.67 | +1.35 | +0.10 |
| 14795-75 | 60.0 | 20.0 | | 20.0 | 0.03 | 1.58 | 5.96 |
| 14795-83 | 60.0 | | 20.0 | 20.0 | +1.66 | 0.05 | 3.95 |
| 14795-85 | 50.0 | | 25.0 | 25.0 | +2.44 | +2.36 | +0.59 |

*A plus sign indicates a weight gain.

Table I-10 ANALYSIS⁵² OF SINTERED CERAMIC TERNARY BODIES AFTER DOUBLE VACUUM SINTERING AND AFTER HEATING IN AIR AT 1750°C FOR 20 HR

| Identification | Intended composition, mole % | | | | Analysis, mole % | | | | | | | |
|----------------|------------------------------|--------------------------------|-------------------------------|-----|-------------------------|--------------------------------|-------------------------------|------|----------------------------|--------------------------------|-------------------------------|------|
| | UO ₂ | La ₂ O ₃ | Y ₂ O ₃ | CaO | After vacuum sintering* | | | | After air firing at 1750°C | | | |
| | | | | | UO ₂ | La ₂ O ₃ | Y ₂ O ₃ | CaO | UO ₂ | La ₂ O ₃ | Y ₂ O ₃ | CaO |
| 14795-67 | 50 | 25 | | 25 | 54.5 | 25.6 | | 19.9 | 54.4 | 25.2 | | 20.4 |
| 14795-75 | 60 | 20 | | 20 | 63.1 | 20.0 | | 16.9 | 62.4 | 21.6 | | 16.0 |
| 14795-83 | 60 | | 20 | 20 | 61.9 | | 20.3 | 17.9 | 63.0 | | 20.6 | 16.4 |
| 14795-85 | 50 | | 25 | 25 | 52.4 | | 25.5 | 22.1 | 55.8 | | 24.4 | 19.8 |

*Binary-oxide powder mixtures (UO₂-Y₂O₃ and UO₂-CaO) were first compacted hydrostatically at 100,000 psi. Compacts were vacuum sintered at 1650°C for 3 hr. Resulting compacts were crushed in an argon-filled dry box. Binary oxide powders were mixed, compacted, and then sintered in vacuum at 1650°C for 4 hr.

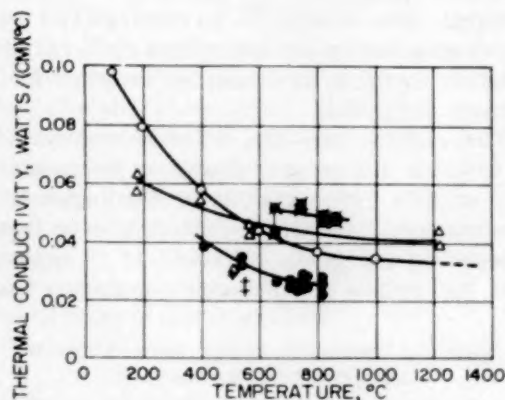


Figure 12—Thermal conductivity of UO₂ plus additives compared to UO₂.⁵³ ○, UO₂. +, UO₂ + 0.25 mole % Nb₂O₅. ■, UO₂ + 4 mole % Y₂O₃. △, UO₂ + 1 mole % Y₂O₃. ●, UO₂ + 0.25 mole % Y₂O₃.

limited to 0.25 mole % to avoid two-phase systems. Four mole % Y₂O₃ increases the thermal conductivity by 25 to 40 per cent between 650 and 850°C. Nb₂O₅ lowers the thermal conductivity. These additives have little effect on the corrosion resistance of the UO₂ in steam.

Hanford irradiation studies⁵⁴ of loose UO₂ powder (4.0 g/cm³) in Zircaloy-2 cans are con-

tinuing. Relocation of the fuel occurs, and the fuel is not moved by tapping or bouncing the can. Sintering produces dense centerline cores (85 per cent of the theoretical density). Radial columnar grains extend from the central high-density cores to cylindrical cracks near the can wall. Solidification from the melt is not necessary for the formation of these columnar grains.

The PuO₂-UO₂ system is also being studied.⁵⁵ Single-phase samples of the two compounds (PuO₂ and UO₂) have been used in a phase study. The liquidus line is simple, and no deviations from uniformity are observed. The lattice parameter of melted PuO₂ is 5.417 Å, compared to 5.396 Å for the as-received powder. There is some evidence for loss of oxygen on melting. Diffraction studies show no evidence of suboxides. PuO₂-UO₂ powder⁵⁶ is also being irradiated in a can as part of the work reported above.⁵⁴

Fueled glass fibers, alone or in a metal matrix, are being studied. A fiber mat may find application in chemonuclear reactors. Harteck and Cyphers⁵⁷ describe potential applications of fueled glass fibers in power reactors. Clevite Corporation⁵⁸ reports work on the fabrication of

aluminum containing aluminum-coated fueled fibers. (W. S. Diethorn)

Fabrication of Uranium Oxides

In previously reported work, Olin Mathieson reported that nitrogen sintering of commercial UO_2 powders containing excess oxygen gave high-density, nearly stoichiometric UO_2 . Later results⁵⁹ suggested that in these experiments the presence of as little as 0.3 wt.% hydrogen impurity in the nitrogen may have contributed to the oxygen removal. Specimens sintered in purer nitrogen did not attain stoichiometry.

It also was observed that, although oxidizing UO_2 powders in air prior to sintering tended to increase the sintered density of all the commercial UO_2 powders used, oxidation did not make it possible to sinter all the powders greater than 95 per cent of the theoretical density.

Du Pont⁶⁰ is investigating explosive forming of "PWR" grade UO_2 to 90 per cent of the theoretical density. Rod up to $\frac{1}{2}$ in. in diameter and 18 in. long, clad in mild steel, was formed. Efforts to use stainless steel or Zircaloy tubing have been less successful because of cladding splitting.

Battelle²⁵ has been working on the growth of UO_2 crystals from the vapor phase, using the reaction UF_4 (gas) + $2\text{H}_2\text{O}$ (gas) \rightarrow UO_2 (gas) + 4HF (gas). In a 100-hr run, several single crystals ranging in size from 6 to 10 mm in length were grown, as well as a large quantity of small cube-shaped crystals about 1 mm³.

(H. D. Sheets)

Properties of Refractory Fuel

Other Than Uranium Oxides

The increasing interest in the use of uranium monocarbide fuel for high-temperature reactors has led to three comprehensive reviews of the literature on the uranium-carbon system.⁶¹⁻⁶³ The first review discusses the published phase diagrams, the various compounds, and some of the reactions of the carbides with uranium oxides, water, and various corrosive media. The second literature survey discusses the chemical reactivity of UC with various groups of elements at high temperature.⁶² The third literature review⁶³ is an annotated bibliography of published work on the uranium-carbon system. A number of reports of studies on UC became available within recent months.

The irradiation behavior of UC is being evaluated at Battelle^{39,64} and at Atomic International. Examinations of irradiated specimens reveal small (2 per cent) volume expansion during the initial exposure, with no further swelling during additional burnup. Fuel specimens of UC tend to crack into large pieces during irradiation, with only a slight amount of spalling. Changes observed in the microstructure indicate that residual UC_2 in UC specimens disproportionates into UC plus carbon during irradiation at 1200°F. Battelle³⁹ has reported an improvement in the strength of UC through the addition of impurity metal and metal carbides. The compressive and transverse rupture strength of UC was increased by a factor of 3 by an addition of 5 wt.% molybdenum, niobium, titanium, or vanadium to arc melts of UC.

Uranium diffusion into UC has been studied at Battelle³⁹ by heating sandwich specimens of U^{235} -UC in the temperature range of 1800 to 2100°C. The diffusion coefficients obtained were in the range of 1×10^{-8} to 1×10^{-6} cm²/sec.

A Union Carbide report⁶⁵ indicates that UC does not react with niobium in 100 hr at 1100°C in argon, suggesting the possible use of niobium for cladding UC fuel elements.

Parth and Pemsler⁶⁶ investigated the tantalum-uranium-carbon system by X-ray diffraction methods and found no evidence for a ternary compound.

The sulfides, selenide, telluride, and carbide of uranium are being evaluated as thermoelectric nuclear fuel materials at Westinghouse.⁶⁷ Preliminary electrical measurements on these compounds are given in Table I-11. It appears that the sulfide and selenide compounds have

Table I-11 THERMOELECTRIC PROPERTIES OF NUCLEAR FUEL MATERIALS⁶⁷

| Compound* | Temp., °C | | Seebeck coefficient, $\mu\text{V}/^\circ\text{C}$ | Resistivity, ohm-cm at 25°C |
|------------------------------|--------------|---------------|---|-----------------------------|
| | Hot junction | Cold junction | | |
| US_x (1) | 45 | 29 | -67 | 0.016 |
| US_x (4) | 77 | 34 | -305 | 0.070 |
| USe_x | 62 | 39 | -171 | 0.019 |
| UTe_x | 102 | 29 | -1.4 | 5.1×10^{-4} |
| UC_x (1) | 93 | 30 | +43 | 1.9×10^{-4} |
| UC_x (2) | 135 | 53 | +22 | 1.03×10^{-4} |
| $\text{UTe}_x + \text{US}_x$ | 151 | 87 | +140 | 0.070 |

*Compositions are not given since the analyses are presently in progress. Numbers in parentheses are specimen numbers.

good possibilities for thermoelectric materials. However, owing to reaction with contact material and to cracking or disintegration of hot-pressed pellets, difficulties have been encountered at elevated temperatures. Measurements were made on US_x-UTe_x pellets up to 810°C , but the pellets failed in tests run at 900°C . Pre- and postirradiation measurements of the electrical properties of $Na_{0.05}U_{0.95}O_2$ have indicated no significant change in this material for irradiations of 1.4×10^{19} nvt.

Refractory plutonium materials are being evaluated for use as high-temperature fuel elements. The reaction and sintering characteristic of PuO_2 mixed with MgO , Al_2O_3 , ZrO_2 , and stabilized ZrO_2 is being investigated at Hanford.^{37, 68} A discontinuity in the thermal expansion of PuO_2 occurs between 600 and 700°C when heated in vacuum but is not observed up to 1000°C when the oxide is heated in a helium atmosphere. The discontinuity has been attributed to evaporation. The reaction of PuO_2 with carbon is reported⁶⁸ to start at 1700°C . Pu_2C_3 is formed with no retention of oxygen in the carbide lattice when PuO_2 is reacted with carbon at 1800°C . The melting point of Pu_2C_3 was found to be 2100°C . The PuC phase diagram was deduced from X-ray diffraction, metallographic, and thermal analyses by Mulford et al.⁶⁹ Four carbide phases (Pu_3C_2 , PuC , Pu_2C_3 , and PuC_2) have been shown to exist. The Pu_3C_2 phase decomposes to Pu and PuC at temperatures above 575°C . PuC_2 appears to be unstable below 1750°C , decomposing into Pu_2C_3 plus carbon. PuC melts at 1654°C , whereas Pu_2C_3 melts at 2050°C . The melting point of PuC is 630°C at the eutectic composition.

(D. A. Vaughan)

Fabrication of Ceramic Fuels

Other Than Uranium Oxides

The preparation and fabrication of UC are being studied extensively at a number of laboratories. Investigations of arc melting both as a method of preparing UC and as a means of fabricating specimens by casting the molten carbide are continuing at Olin Mathieson,⁵⁹ Atomics International,⁷⁰ and Battelle (references 25, 39, 71, and 72). The reaction of uranium metal or uranium hydride with methane or propane to produce a UC powder is under study at Olin Mathieson,⁵⁹ Battelle (references 25, 39, 71, and 72), Carborundum,⁷³ and Los Alamos.⁷⁴ The optimum temperature⁵⁹ for reaction with

methane appears to be about 700°C . Reduction of uranium oxides^{70, 73, 74} or ammonium diuranate^{75, 76} with carbon offers still another method of preparing the monocarbide.

In addition to melting and casting as a means of fabricating UC, sintering of cold-pressed specimens (references 25, 39, 59, 71, 72, 74, 75, 77, and 78) and hot pressing^{76, 78, 80} continue to receive attention. Uranium carbide compositions slightly deficient in carbon appear to have optimum sintering characteristics.⁵⁹ Relatively wide variations in the initial particle size, the forming pressure, and the sintering temperature and time were found to have little effect on the bulk density, grain size, and porosity of the resulting sintered UC.⁷⁸ In hot-pressing procedures the use of aluminum nitride as a die material, in place of the graphite commonly used, minimized reaction between the UC and the die.⁷⁷ A hot press in which the graphite die is heated by electric-resistance heating rather than by induction heating enables rapid production of hot-pressed specimens.⁸⁰ The complete cycle of heating to 2500°C , applying pressure, and cooling required only 10 min with the resistance-heated die.

Other uranium compounds being studied as potential nuclear fuels include UN and uranium silicides. The preparation of UN by nitriding a mixture of UO_2 and carbon or the more common method of nitriding uranium metal is under study at Carborundum.^{74, 76} In attempting to prepare U_3Si_2 by hot pressing a stoichiometric mixture of uranium and silicon metals, mixtures of the various uranium silicides were produced.⁷⁵ The compound was successfully prepared by quenching small melts of uranium and silicon.⁷⁶ A program on the fabrication of UC-PuC mixed fuels has been started as a joint effort of Nuclear Development Corporation and Carborundum.^{77, 81}

(M. J. Snyder)

Mechanism of Corrosion

of Fuel Alloys

Hydrogen in Corrosion Films. Work performed at the Imperial College, London,⁸² is the basis for the proposal that hydrogen or hydrogen ions in the oxide film influence the corrosion rates of such metals as uranium, aluminum, and manganese. It is predicted that depolarization of hydrogen before its entry into the film or a high solubility for hydrogen in the base metal

would lessen the hydrogen content in the oxide film and hence improve corrosion resistance. Examples of treatments that result in decreased corrosion rates are cited as (1) the use of nickel and iron in aluminum alloys to depolarize hydrogen⁸³ and (2) the increase of zirconium content or gamma stabilization in uranium-zirconium alloys to obtain greater hydrogen solubility.⁸⁴ On the other hand, it should be noted that Bettis has reported similar corrosion behavior in water and oxygen for zirconium-uranium alloys and Zircaloy-2, which led to the conclusion that hydrogen had no effect on the corrosion kinetics of these alloys.^{85,86} (W. E. Berry)

Basic Studies of Irradiation Effects in Fuel Materials

Nonmetallic Fuels

Hanford research^{6,68,87} on the effects of thermal-neutron irradiation on thin UO_2 and ThO_2 films has continued. All reported effects from exposures to 4.4×10^{19} nvt have been ascribed to fission fragment-lattice interaction. Transmission electron diffractometry has revealed that the originally crystalline UO_2 film, 200 Å thick, is transformed by increasing irradiation to a distorted crystalline structure until at about 4×10^{19} nvt the structure becomes amorphous. Additional exposure to 4.4×10^{19} nvt apparently causes recrystallization of the amorphous structure, and sharp diffraction patterns are re-formed. Fission-fragment tracks appearing on electron micrographs have a length distribution which would be expected on the basis of geometrical considerations. Similar effects have been observed in irradiated ThO_2 thin films. Structural features of fission-fragment tracks indicate that, although the ambient temperature of irradiation was only 45°C, sufficient heating occurred on the microscale to cause reaction of the oxide with residual O_2 in the irradiation capsule.

Irradiation exposures to 8×10^{18} thermal nvt were sufficient to sublime microscopic UO_2 particles enclosed in air-filled capsules.⁶⁸ Similar exposures in evacuated capsules did not result in specimen sublimation.

Westinghouse⁵¹ has also been concerned with irradiation effects on crystallographic features in fuel materials. The compounds Al_2O_3 , ZrSiO_4 , ZrO_2 , U_3O_8 , UO_2 , and CaO and their mixtures

were exposed to thermal-neutron doses of 10^{18} to 10^{19} nvt. All have a high degree of stability to neutron irradiation. However, with the exception of UO_2 , the compounds and their mixtures are severely affected by fission-fragment interaction. Al_2O_3 and ZrSiO_4 matrix materials containing dispersions of UO_2 became amorphous. Grain boundaries disappeared, and X-ray diffraction patterns became diffuse. Similar effects occurred in the mixture ZrO_2 -13 wt.% CaO -17 wt.% UO_2 . The U_3O_8 crystal structure was destroyed after a thermal-neutron exposure of 1×10^{16} nvt. Only slight line broadening was noticed in the UO_2 X-ray diffraction. The solid solution ZrO_2 -80 wt.% UO_2 was unaffected by a neutron exposure of about 10^{19} nvt. The general conclusion derived from these experiments is that only the face-centered cubic structure with its relative isotropy is resistant to fission-fragment damage, whereas the noncubic anisotropic structures are severely affected by fission-fragment interaction. The mechanism for such extensive effects is still not apparent.

Research into the release of fission gases was continued at Westinghouse.⁸⁸ Two materials were considered: a $\text{UO}_{2.002}$ specimen prepared from uranyl nitrate and a UO_2 specimen obtained by crushing sintered UO_2 pellets to -200 +325-mesh size. In postirradiation gas-release experiments in the range 900 to 1500°C, the effects of progressive sintering during heating were of interest. Control experiments showed that surface areas decreased by a factor of about 5 during a 22-hr heat-treatment at 1400°C in a hydrogen atmosphere. With allowance for the reduction of the gas-releasing surface during gas release and also for the depletion of fission gas in the small-particle fraction, the equations for the diffusion coefficient of Kr⁸⁵ in the uranyl nitrate UO_2 and in the crushed sintered UO_2 , respectively, are as follows:

$$D = 2.65 \times 10^{-4} \exp(-65,500/RT)$$

$$D = 4.9 \times 10^{-4} \exp(-73,800/RT)$$

Similar postirradiation fission-gas-release experiments continued at Battelle.^{25,71} Single-crystal platelets of fused natural UO_2 were heated in a flowing atmosphere of purified helium at 1205 and 1245°C after irradiation exposures of 5.8×10^{17} nvt and 8.6×10^{17} nvt thermal. The reduction of data to diffusion coefficients for xenon and krypton isotopes is not complete.

A mathematical evaluation at Knolls⁸⁹ dealt with the effect of gas bubbles on fuel-element swelling. In a fuel assumed to offer no resistance to swelling other than surface tension, the fractional swelling is inversely proportional to the square root of the number of bubbles. The size distribution of the bubbles, however, is not a factor unless the bubble becomes so large that only a few of the bubbles are very large.

(F. A. Rough)

Basic Effects of Irradiation upon Fuels:

Metallic Fuels

A number of reviews and general papers appeared in the literature within the past few months which were concerned primarily with the theory of damage in metals or metallic fuels. Brooks⁹⁰ describes the mechanisms of radiation damage and some of the resulting effects. The discussion is divided into three categories: (1) mechanisms of damage production, (2) nature and mobility of the imperfections produced, and (3) effect of the imperfections on the measurement of the properties of the solid. The review is concerned primarily with metals and semiconductors.

Lillie⁹¹ has reviewed the experimental data and mechanisms for essentially all nuclear and basic cases involving the production of gases in solids. The subjects discussed in this review are: (1) alpha bombardment of copper; (2) gamma irradiation of Lucite; (3) neutron irradiation of lithium fluoride; (4) neutron irradiation of magnesium-lithium and aluminum-lithium; (5) neutron irradiation of beryllium metal; (6) neutron irradiation of boron-containing materials; and (7) uranium fission in metal, dispersion, and ceramic fuels.

General articles also appeared in the literature, including a paper by Bentle⁹² on the relation between preirradiation properties and irradiation swelling and a paper by Bosnak⁹³ on mechanisms and manifestations of irradiation damage in reactor materials.

At Hanford,⁶ fractographic examination of uranium reveals that fracture surfaces of uranium, produced at -196 and -78°C after irradiation at about 200°C to 0.016 at. % burnup, are similar to fracture surfaces of unirradiated materials with one exception. In the irradiated specimen fractured at -78°C , twinning, which is normally observed in the unirradiated material, is absent.

At Advance Technology Corporation laboratories,⁹⁴ the feasibility of improving swelling resistance in metallic uranium by increasing the dislocation density is under investigation. Since this research has just begun, results are limited to metallurgical developments preparatory to the study of irradiation effects. Increasing the density of dislocation is expected to (1) increase the number of sites at which fission-product-gas atoms are "pinned," (2) increase the number of gas-bubble nuclei, and (3) increase mechanical strength.

(F. A. Rough)

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Graphite

Samples of candidate graphites for the New Production Reactor (NPR) were irradiated at Hanford¹ to 2500 Mwd/at at 550 to 600°C. [One Mwd/at is the amount of irradiation received by a sample during the time required for the ton of uranium adjacent to it to generate 1 Mwd of fission energy. For the facility used to obtain the data in Tables II-1 and II-2, this amounts to an exposure of about 6.5×10^{16} (above 1 Mev).] The samples were annealed by heating at 650°C prior to irradiation. The dimensional changes are presented in Table II-1. All the graphites derived from needle coke (GL-10, GL-11, VC, and KC) exhibited expansion in the transverse direction, whereas those from other cokes (CSF and TSGBF) contracted.

Several high-density graphites prepared by pressure forming and baking processes were irradiated under similar conditions. Length changes are summarized in Table II-2. These specimens will be reirradiated to determine whether the high initial contraction rates continue at higher exposures.

Several less permeable types of graphite were irradiated in the Engineering Test Reactor (ETR) to 1×10^{20} nvt (>1 Mev) at 400 to 600°C,² with the results shown in Table II-3.

Constant-load tensile-creep properties of graphite were measured at 1650 to 2900°C for

periods of time up to 8 hr at the California Institute of Technology Jet Propulsion Laboratory.³ Specimens had a gauge section $\frac{1}{4}$ in. in diameter by 1 in. long. They were heated in helium by a graphite heater. Creep rates of 10^{-8}

Table II-1 IRRADIATION¹ OF NPR CANDIDATE GRAPHITES TO 2500 MWD/AT AT 550 TO 600°C

| Graphite type | Length change, % | |
|---------------|------------------|----------|
| | Transverse | Parallel |
| GL-10 | +0.022 | |
| GL-11 | +0.015 | +0.003 |
| VC | +0.006 | -0.019 |
| KC | +0.025 | -0.017 |
| CSF | -0.005 | -0.022 |
| TSGBF | -0.008 | -0.012 |

Table II-2 IRRADIATION¹ OF HIGH-DENSITY GRAPHITE TO 2500 MWD/AT AT 550 TO 600°C

| Type | Approximate density, g/cm ³ | Length change, % | |
|----------------------|--|------------------|----------|
| | | Transverse | Parallel |
| NC-1 (unimpregnated) | 1.81 | -0.085 | -0.091 |
| NC-2 (impregnated) | 1.90 | -0.128 | -0.120 |
| NC-3 (unimpregnated) | 1.83 | -0.095 | -0.092 |
| NC-4 (impregnated) | 1.92 | -0.087 | -0.087 |

Table II-3 IRRADIATION RESULTS FOR LESS PERMEABLE GRAPHITES²

| Sample type | Length change, % | Weight change, % | Original C_0 , A | Change in C_0 , A | Original L_0 , A | Change in L_0 , A | Density, g/cm ³ | CTE* |
|-------------|------------------|------------------|--------------------|---------------------|--------------------|---------------------|----------------------------|------|
| GLC type A† | | -0.08 | 6.734 | +0.001 | 420 | -105 | 1.78 | 1.08 |
| GEC-3‡ | -0.05 | -0.11 | 6.734 | +0.005 | 265 | +5 | 1.74 | 2.75 |
| HS-1§ | -0.05 | -0.52 | 6.725 | +0.019 | 345 | -95 | 1.79 | 3.67 |
| GLC type P† | -0.10 | -0.27 | 6.727 | +0.013 | 380 | -75 | 1.74 | 1.06 |
| HS-4§ | -0.12 | -0.30 | 6.728 | +0.014 | 300 | -53 | 1.81 | 3.20 |
| GEY¶ | -0.40 | -0.99 | 6.719 | +0.012 | 470 | -188 | 1.90 | |

*Ratio of coefficient of thermal-expansion values measured perpendicular and parallel to extrusion axis; these are preirradiation values supplied by General Atomic, source of the graphite samples.

†Great Lakes Carbon.

‡G. E., England.

§Hawker-Siddeley, England.

¶National Carbon.

to 10^{-7} /sec were observed at stresses of 1600 to 4000 psi. Creep rates increased continuously with increasing temperature and did not show a minimum corresponding to the peak in tensile strength at about 2500°C. Creep rates were lower when stress was applied parallel to the grain than they were when stress was transverse. Elongations up to 40 per cent and density decreases up to 18 per cent were observed.

(J. Koretzky)

Beryllium

Several extensive bibliographies⁴⁻⁸ on beryllium have been received in recent months. Together, they cover all phases of current research on this metal. One of these items⁷ is a convenient reference source for information on the physical, mechanical, and chemical properties of beryllium.

A description of a new plant that is expected to process about 7 tons of wrought beryllium per year for the United Kingdom Atomic Energy Authority (UKAEA) appeared in the open literature.⁹⁻¹² Beryllium fabricating methods that will be used there are also described.¹³ Ladd and Albrecht¹⁴ of Brush Beryllium Company reported on the fabrication of reflector parts for the Br-2 reactor for the Center for the Study of Nuclear Energy (CEN) at Mol, Belgium. The reactor was designed cooperatively by Nuclear Development Corporation of America and the CEN. Additional evidence of international cooperation is seen in the announcement¹⁰ that Chesterfield Tube Company of Great Britain has agreed to furnish Superior Tube Company of Norristown, Pa., with the techniques of drawing and finishing beryllium tubes developed by the former's parent organization, Tube Investments Ltd. Current French interest in beryllium is indicated by two publications^{15,16} on properties, fabrication, and utilization of beryllium.

Fabrication

The experience gained in producing beryllium sheet with a certain amount of bidimensional ductility and in fabricating this material into a load-bearing structural assembly by forming and riveting was described in the final report of Nuclear Metals, Inc., to the Martin Company.¹⁷ The effects of several processing variables on strength and ductility of the sheet were evalu-

ated. The sheet finally produced had the properties shown in Table II-4.

The feasibility of extruding bare beryllium shapes was reported previously. This work was continued, and 10- to 12-ft lengths of U-shaped channel have been produced. Efforts to develop a commercial process to make 20-ft-long extrusions are continuing.^{18,19}

Table II-4 MECHANICAL PROPERTIES OF UPSET BERYLLIUM SHEET¹⁷

[Reduction in Area, 8:1; Upsetting Temperature, 1900°F; Vacuum Annealed at 1390°F (750°C) for 1 Hr]

| Property | Value |
|------------------------------------|-------------|
| Modulus of elasticity, 10^6 psi | 42 |
| Yield strength (0.05% offset), psi | 31,000 |
| Engineering tensile strength, psi | 60,000 |
| Uniaxial elongation, % | 13 |
| Biaxial elongation, % | 1.2 to 1.9* |

*Dependent on testing technique.

Extensive tensile testing, at both room and elevated temperature, showed that beryllium sheets rolled from vacuum-hot-pressed billets made from -325-mesh powder were stronger than similar sheets made from -200-mesh powder. The added strength of the -325-mesh powder came from a higher oxide content, unavoidable in finer grinding. However, the higher oxide made the sheets more fragile and difficult to roll, and therefore this approach is being abandoned.²⁰

Beryllium reacts at elevated temperatures with most elements to form brittle intermetallics, a characteristic which makes it difficult to clad successfully. However, it apparently forms no compounds with silicon. Ductile intermetallics are formed with yttrium and aluminum; hence it may be possible to incorporate yttrium, aluminum, or silicon in areas where undesirable metallics are likely to form. Beryllium also could be coated with aluminum by spray or dip methods. Subsequent oxidation of the aluminum to form a thin layer of Al_2O_3 would prevent intermetallic reactions between beryllium and the cladding, and, at the same time, it would form a stable coating compatible with the beryllium.

Beryllium, it was reasoned,^{21,22} had too low a ductility to be drawn into wire at room temperature and too low a cohesive strength in tension to be drawn at the high temperatures that are commonly used for hot rolling. However, in the

warm-working temperature range of 600 to 1000° F, the ductility and strength are both good. Also, in this range the oxide formed is tenacious and protective. Following this reasoning, about 200 ft of 0.063-in.-diameter wire was successfully drawn from 0.254-in.-diameter rod, using MoS₂-epoxy resin as a lubricant. Two intermediate anneals of 30 min at 1480° F were used. The final wire, tested at 800° F, had a yield strength of 52,000 psi and showed a reduction in area at the fracture of 17.9 per cent.

The problems of joining beryllium are not as insurmountable as was thought originally. Although beryllium must be regarded as a crack-sensitive material, satisfactory conditions for fusion welds have been obtained in all joint designs attempted except in sheet butt welds. Brazing of beryllium to itself and other metals has been successfully done using gas-torch and furnace heating with flux additions. Fluxless brazing may be employed using inert-gas-shielded arc welding methods with consumable-wire electrodes. Pressure and resistance welding methods produce sound homogeneous bonds, but they are limited to those applications where the design permits the parent beryllium to withstand the high deformation required.²³

The Wright Air Development Center and Avco Manufacturing Corporation jointly sponsored a program on brazing and welding of beryllium. A final report²⁴ on the company-sponsored part of the program has been made public. It is reported that beryllium-rich silver alloys with about 20 at.% silver were ductile and could be cold rolled to a reduction in thickness of 50 per cent or more. Strong brazed joints of beryllium to itself and to some other metals can be made with silver if the silver is preplaced in the joints. On heating, the silver alloys rapidly wet the beryllium, but neither silver nor the silver-beryllium alloy flows readily on beryllium. On the part of the program sponsored by the Air Force, factors affecting the room-temperature tensile properties of aluminum-12 wt.% silicon and silver braze welds were investigated; the mechanism of tearing in fusion welds in beryllium was discussed, and the effects of a number of variables were determined on the strength of bonds in beryllium formed by solid-state welding.²⁵

Meanwhile, workers at Oak Ridge reported²⁶ that beryllium joints made by the inert-gas-shielded arc welding process showed considerable porosity that could be minimized by the use

of alternating current and a copper chill ring. This group also found that an experimental brazing alloy, 49 wt.% titanium-49 wt.% copper-2 wt.% beryllium, exhibited excellent flowing and wetting properties on beryllium. Leaktight joints could be formed with this alloy, and also by diffusion bonding.

The Air Force has established a program²⁷ at Standard Pressed Steel Company to determine the feasibility of fabricating threaded fasteners from beryllium. The company found that the first experimental bolts exhibited some properties that exceeded those of either steel or titanium on a strength-to-weight basis. A detailed report was made of the inspection procedures that were used and of the results of an extensive mechanical-testing program.

At Oak Ridge it has been found²⁶ that inclusions of mild steel left on the inside of beryllium tubing during the extrusion operation interfere with radiographic inspection and that they must be removed. Ultrasonic inspection revealed one crack, missed by X-ray examination, which extended nearly through a tube wall from the inside.

In the tubes from Nuclear Metals, Inc., the oxide consists of small particles distributed randomly regardless of grain boundaries, forming loose stringers in the extrusion direction.

Physical and Mechanical Properties

One of the major difficulties standing in the way of wider application of beryllium is its brittleness. Much of the research in progress on this metal, therefore, is designed either to find a way to improve its ductility or simply to understand the reason it is brittle. The Navy is sponsoring one program²⁸ to try to make ductile alloys by liquid-phase sintering of Be-Ag-X and Be-Al-X alloy systems, with the objective of producing an envelope type of microstructure. The Navy also is sponsoring an attempt to make very pure beryllium in the hope that it will be ductile. Summarizing a literature review made earlier, the current report²⁹ concludes that elimination of impurities can extend the range of plastic deformation of beryllium if the impurities (1) limit extensive dislocation motion on the basal plane or (2) act as nuclei for cracks that form during kinking.

Following an extensive survey of the literature concerning methods of controlling textures in sheet metals, an experimental program³⁰ was

laid out with the hope that it may make possible the production of isotropic, ductile beryllium sheet.

Continuing a work of which the first part was published in 1955, Garber and coworkers³¹ presented an important article on the crystallographic and mechanical characteristics of various types of plasticity and decomposition in beryllium monocrystals in the temperature range from -253 to $+800^{\circ}\text{C}$. It was confirmed that slipping along the (0001) plane in the $[11\bar{2}0]$ direction is the basic type of plastic deformation in beryllium from -196 to $+800^{\circ}\text{C}$. Erratic shifting along the basal slipband causes microcracking along the prism and pyramid planes.

The AEC is sponsoring a two-pronged attack on the beryllium ductility program.^{32,33} A body-centered cubic phase (β) exists at temperatures very close to the melting point. If the beta phase could be retained at room temperature, presumably it would be more ductile than the close-packed hexagonal phase that is stable at all lower temperatures. Gelles and Pickett found that nickel appears to have a marked effect on expanding the beta-phase region. The data obtained on alloys with 1.0 and 5.0 at.% nickel supported the phase diagram for the beryllium-nickel system that had been proposed by Gelles et al.³⁵ On another front, a project known as the Beryllium Fracture Program, X-ray rotation patterns of beryllium single crystals were examined. It was found that particles rich in iron, manganese, and chromium precipitate on aging at elevated temperatures. Quenching from temperatures as high as 1200°C did not eliminate the precipitated particles. Commercial sintered polycrystalline beryllium was solution-treated for 1 hr at 1100°C and was either quenched or slowly cooled to room temperature. The quenched samples were aged at either 400 or 600°C for various lengths of time. None of the unaged specimens showed a yield point in tensile tests. All the specimens aged at 400°C for 2 hr or more showed definite yield points, whereas those aged at 600°C developed a yield point only after a much longer aging period.

An attempt was made³⁵ to correlate the structure-insensitive physical properties of beryllium on the basis of the theory of solids. The author proposed an explanation of the decrease of lattice-parameters ratio with temperature based on the Herring-Hill calculations of the wave functions for beryllium.

The solid-solubility limits of iron and nickel in beryllium were determined between 850 and 1200°C . The maximum solubility of iron was 0.92 ± 0.02 at.% (5.46 wt.%) at 1225°C . The maximum solubility of nickel was 4.93 ± 0.01 at.% (25.2 wt.%) at the eutectoid temperature, 1065°C . A high-temperature phase (β) was found in the beryllium-nickel system at approximately 8 at.% (36 wt.%) nickel and was reported³⁴ for the first time.

Curves showing the relation between true stress and plastic strain for extruded and cross-rolled beryllium sheet have been published.³⁶

Chemical and Thermal Properties

A British patent³⁷ described the preparation of beryllium alloys containing plutonium, uranium, or actinium. Powdered beryllium was mixed with UF_4 , PuF_4 , or AcCl_3 and heated *in vacuo* to reduce the halide, distill the BeF_2 or BeCl_2 formed, and alloy the remaining metals.

A proprietary method, "Dalic," for depositing gold on beryllium, either directly or over a preliminary nickel plate, was found unacceptable at the University of California Lawrence Radiation Laboratory (UCLRL) because of voids or pinholes. Satisfactory gold plates were obtained from cyanide baths directly on beryllium.³⁸ It was also noted that conflicting results continue to be accrued on the behavior of porous beryllium in HNO_3 -Pu solutions. It is believed that the variation may be due to occluded metals in the beryllium or to other materials in the test assembly.

A detailed description of the spectrographic technique used by Avco Manufacturing Corporation for the analysis of beryllium in air samples has been published.³⁹ The method is said to be sensitive to a minimum of $0.05 \mu\text{g}$ of beryllium with a relative error of less than 15 per cent of the amount present in the range from $\frac{1}{2}$ to $2 \mu\text{g}$.

The mechanism of vaporization of beryllium and the source of oxygen in the distilled metal were analyzed.⁴⁰ It was shown experimentally that beryllium evaporates from the solid phase in the form of Be_2 dimer and from the liquid phase as Be monomer. The reaction $\text{Be} + \text{BeO} \rightarrow \text{Be}_2\text{O}$ takes place at 1400°C and up, with the product Be_2O remaining stable at ordinary temperatures. The lower beryllium oxide has a higher volatility than BeO and is one of the

sources of oxygen appearing during BeO distillation in the crucible.

The nuclear and mechanical properties of beryllium and its oxide were tabulated⁴¹ and compared with those of zirconium, magnesium, and aluminum to show that beryllium is superior for use as a canning material and that the advantage is greater the higher the temperature (from 20 to 500°C). Brief information was also given on processing, plastic deformation, and physical, chemical, and mechanical properties of beryllium.

The thermal conductivity of beryllium was found to depend on the orientation with respect to the pressing axis. The thermal conductivity is higher in the direction perpendicular to the direction of pressing than in the parallel direction (unlike graphite). It is expected that improving the purity of the beryllium would increase the thermal conductivity appreciably.

At Oak Ridge, studies were initiated to determine the maximum permissible temperature for the exposure of beryllium in various gases.²⁶ The reaction $\text{Be} + \text{CO}_2 \rightarrow \text{BeO} + \text{CO}$ appeared to take place during exposure of beryllium to CO_2 at 700°C with an initial pressure of 200 mm Hg.

Significant quantities of gas are produced in beryllium as a result of nuclear reactions on exposure to a fast neutron (>1 Mev) dose of 10^{22} neutrons/cm². It has been reported that the volume increase in beryllium containing 23 cm³ of helium per cubic centimeter of beryllium would exceed 1 per cent. Studies²⁶ are under way to determine the effects of void formation on the ability of a beryllium cladding material to retain fission-product gases and on its resistance to failure under thermal and mechanical stresses.

C. E. Ellis and W. Evans [Report CRMet-864 (AECL-898)] showed that, if an arbitrary limit of 1 per cent swelling is adopted, this would not be exceeded until the temperature is above 600°C. Experiments are being conducted on the irradiation of beryllium of different degrees of purity to determine the effect of inclusions on the nucleation of gas bubbles. A paper describing the elevated-temperature irradiation technique is being published in *Nuclear Instruments & Methods*, Volume 6.

Miscellaneous

Several papers⁴²⁻⁴⁴ on the control of beryllium hazards have appeared. Important points in the

design and satisfactory operation of a beryllium workshop are outlined, and the procedures used for control at specific installations are described. (W. Hodge)

Beryllium Compounds

Reactions of water vapor with BeO, BeO·Al₂O₃, and BeO·3Al₂O₃ were studied at Atomics International in the 1300 to 1570°C temperature range.⁴⁵ Results indicate that the addition of alumina to beryllia does not decrease the volatility of the latter sufficiently to reduce health hazards or corrosion rates associated with the use of BeO in contact with moist air at high temperatures to acceptable levels.

(J. Koretzky)

Solid Hydrides

The Proceedings of the Second Metal Hydride Conference were compiled by Denver Research Institute.⁴⁶ This publication comprises 20 papers covering various aspects of the technology of zirconium and yttrium hydrides.

Zirconium Hydride. The specific heat of ZrH_{1.6} has been reported by Atomics International.⁴⁷ The values range from 0.11 cal/(g)(°C) at 0°C to 0.15 cal/(g)(°C) at 250°C. They have also measured electrical resistivities and Hall coefficients for zirconium hydride with hydrogen-to-metal ratios from 1.54 to 1.96 over the temperature range 1.1 to 300°K.⁴⁸ At room temperature the resistivity is 24.7 μohm-cm for the composition ZrH_{1.96}. The Hall coefficients indicate majority hole conduction for the face-centered cubic structure and majority electron conduction for the face-centered tetragonal type.

The diffusion of hydrogen in the delta phase of zirconium hydride has been measured at Battelle⁴⁹ using permeation techniques. The data were obtained for compositions from 61 to 65 at.% in the temperature range 500 to 750°C. Diffusion coefficients obtained by the time-lag method showed that there is no variation of coefficient with hydrogen concentration. Figure 13 shows the change of diffusion coefficient with temperature, as given by the empirical relation $D \text{ (cm}^2\text{/sec)} = 599 \exp (-34,800/RT)$.

A review of the data on hydrogen pickup in zirconium alloys has been prepared by the

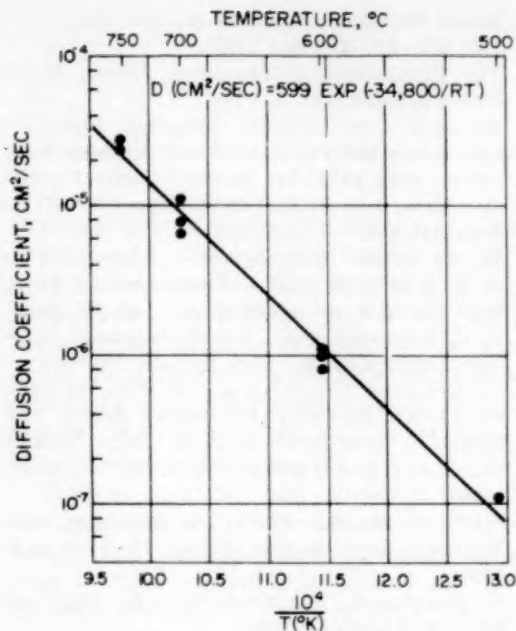


Figure 13—Effect of temperature on the diffusion coefficient of hydrogen in delta-phase zirconium hydride.⁴⁹

Canadians.⁵⁰ The pickup in reactor components such as Zircaloy-2 fuel sheaths has been predicted to be 58 ppm/year with a 10-mil cladding thickness and 19 ppm/year with a 30-mil sheath. For pressure tubes a pickup of only 3.7 ppm/year is predicted. Bettis made experimental studies of the thermal diffusion of hydrogen in Zircaloy-2 in the temperature range 300 to 500°C.

The compatibility of zirconium hydride and carbon dioxide at 1200°F was studied by General Atomic.⁵¹ The $ZrH_{1.5}$ displayed excellent dimensional stability but lost up to 20 wt.% of its hydrogen. The hydrogen loss in 1000 hr was 3 per cent at 800°F, 10 per cent at 1000°F, and 20 per cent at 1200°F. In the case of the two-phase $ZrH_{0.9}$, the material swelled and warped in the 400°F temperature gradient, but it did not lose much hydrogen or corrode severely.

(H. H. Krause)

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Burnable-poison plates are being developed at Bettis,¹ and they consist of wafers of hot-pressed high-density B_4C clad with 15 mils of Zircaloy-4. The cladding is pressed into intimate contact with the B_4C and is clad to Zircaloy spacers either by a gas-pressure-bonding procedure or by copper-diffusion-bonding techniques. Wafers of 50 vol.% B_4C in SiC are also being bonded by these techniques.

General Electric² has developed techniques for pressing and sintering rare-earth oxide wafers that can be welded into stainless-steel cans to form a cruciform control rod for the Dresden reactor; one type of wafer consists of 45 wt.% Gd_2O_3 -45 wt.% Sm_2O_3 (the remaining 10 per cent was not identified), and the other type of wafer is a composite consisting of a layer of Dy_2O_3 sandwiched between two layers of Gd_2O_3 . The compacts are pressed with no binder at pressures of 10,000 to 20,000 psi and sintered at 1650°C for 2 hr. Sintered densities obtained are 93.9 per cent of the theoretical density.

Vallecitos³ is making a survey of borides for use as control materials and has recently released a report on these materials. Some of the data obtained for the rare-earth borides are listed in Table III-1.

Table III-1 STRUCTURE OF RARE-EARTH BORIDES³

| Compound | Lattice constant (cubic structure), A | Density, g/cm ³ | Boron present, g/cm ³ |
|------------------|---|-------------------------------|--|
| EuB ₄ | 4.175 | 4.95 | 1.48 |
| SmB ₄ | 4.129 | 5.07 | 1.53 |
| GdB ₄ | 4.11 | 5.25 | 1.54 |
| DyB ₄ | 4.13 | 5.35 | 1.53 |

General Atomic⁴ is using a flame-spraying technique to coat stainless-steel tubes with rare-earth oxides. An inner graphite core is used to prevent rapid cooling of the stainless steel. Mixtures of samarium-gadolinium oxide and gadolinium oxide have been successfully applied.

Table III-2 HELIUM RELEASE DURING IRRADIATION OF BORIDES IN THE FIRST SERIES OF CAPSULES⁵

| Capsule | Con- tents | Particle size, U. S. mesh | B ¹⁰ burn- up, % | Helium-release data, * % | | |
|---------|------------------|------------------------------------|-----------------------------------|--------------------------|------------------------|-------------------------|
| | | | | As re- ceived† | 1 week at 700°F‡ | 2 weeks at 700°F‡ |
| BMI-7-1 | B | -60 +80 | 20 | <0.01 | 1.0 | 2.4 |
| BMI-7-2 | B | -60 +80 | 64 | 34.1 | 1.2 | 1.3 |
| BMI-7-3 | ZrB ₂ | -30 +60 | 19 | <6.0 | | <0.4 |
| BMI-7-4 | ZrB ₂ | -30 +60 | 41 | <0.1 | 0.02 | <0.03 |
| BMI-7-5 | B ₄ C | -50 +60 | 22 | <0.1 | <0.02 | <0.01 |
| BMI-7-6 | B ₄ C | -50 +60 | 66 | 5.6 | | |

* Percentage of total helium at given burnup.

† Capsule as received from the MTR and punctured at room temperature.

‡ Heated under a vacuum of less than 5 μ Hg pressure.

Battelle⁵ has recently published a report on radiation effects in boron-containing compounds. Elemental boron, boron carbide, and zirconium diboride were irradiated at reactor ambient temperatures (150°F) and elevated temperatures (500 to 700°F) to burnups ranging from 20 to 95 per cent of the B¹⁰ isotope. Zirconium diboride containing both natural and enriched amounts of B¹⁰ was evaluated. The amount of helium released during irradiation was measured, and X-ray diffraction, metallographic, and chemical examinations were performed both before and after irradiation.

Table III-3 HELIUM RELEASE DURING IRRADIATION OF BORIDES IN THE SECOND SERIES OF CAPSULES⁵

| Capsule | Contents* | Irradiation temp., °F | B ¹⁰ burnup, % | He released, % of He produced |
|----------|----------------------|-----------------------------|---------------------------------|-------------------------------------|
| BMI-7-14 | ZrB ₂ (N) | 150 | 67.8 | 0.6 |
| BMI-7-15 | ZrB ₂ (E) | 500 | 33.0 | 57.0 |
| BMI-7-16 | ZrB ₂ (N) | 500 | 84.7 | 5.2 |
| BMI-7-17 | B ₄ C(N) | 500 | 94 | 67† |

* Particle size: -80 +100 mesh. (N) denotes natural boron, and (E) denotes boron enriched in the B¹⁰ isotope.

† This value was calculated from irradiation data obtained from the MTR. All other values were based on lithium analysis of irradiated powder.

Zirconium diboride containing natural boron appeared to be the most promising material from a helium-retention standpoint. None of the materials exhibited excessive particle fragmentation or dimensional changes during the course of the experiments, although there was evidence of temperatures much higher than design conditions. Some of the data on helium release are shown in Tables III-2 and III-3.

(G. W. Cunningham)

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Corrosion

Niobium Oxidation

A recent study in England¹ on the oxidation reactions of niobium in dry and moist air and oxygen at 400 to 1050°C has provided further confirmation that the oxidation rates show a maximum at about 600°C. This study also indicates a minimum in the linear oxidation rate at about 700°C and a negative inflection in the rate-versus-temperature plot at about 850°C. The temperature dependency of the linear oxidation rates was similar in each of the four atmospheres studied. The anomalous behavior in the 600 to 700°C temperature range may be associated with small compositional changes in the pentoxide scale. Analyses indicated that the scale contained more than the stoichiometric amount of oxygen when formed at 450°C but less than the stoichiometric amount when formed at 650°C. The inflection at 850°C may be associated with the transformation of the pentoxide from the low-temperature form to the high-temperature form, as also suggested by Goldschmidt.²

A literature survey³ on the oxidation behavior and development of protective coatings for niobium and niobium-base alloys has been completed by the Defense Metals Information Center. The most oxidation-resistant niobium alloys are those containing chromium-aluminum, iron-aluminum, molybdenum-aluminum, titanium-aluminum, titanium-chromium, and chromium-cobalt (in order of decreasing oxidation resistance). Alloys in the first three systems are superior to the popular Fecral alloy in high-temperature oxidation resistance, but compositions of the better alloys are in ranges where brittle intermetallics are expected to form. The most effective coatings for niobium include a duplex LM-5 coating and a proprietary nickel-chromium coating. These coatings provide protection for 1000 hr at 2100 and 2200°F, respectively.

The Naval Research Laboratory⁴ has recently published information on zinc-coating of niobium. The zinc is applied by dipping or vapor deposition, followed by annealing at 1600°F in

air to produce niobium-zinc intermetallics on the surface. In static oxidation, these coatings are protective for several hundred hours at 1800°F and for a few hours at 2200°F. The self-healing qualities of these coatings are exceedingly good. Defects as large as 1/4 in. wide heal rapidly at 1800°F.

Battelle^{5,6} is continuing its investigation of hot-water and steam corrosion of niobium and niobium alloys. The most attractive alloy from this investigation, based on cross section, corrosion resistance, and 1200°F mechanical properties, is niobium-12 at.% vanadium. Purity of the niobium base has little effect on alloy corrosion resistance, as shown by controlled carbon and oxygen additions to niobium-vanadium ternary alloys.

(W. D. Klopp)

Zirconium Corrosion

Metallographic studies have been conducted at Bettis⁷ to determine the relation between metal structure and the presence or absence of fleck type markings in the black-oxide films developed on Zircaloy-2 during exposure to high-temperature water. The data suggest that the amount and distribution of the intermetallic precipitates are directly related to the formation of a discontinuous oxide film. For those Zircaloy-2 materials which did not exhibit flecking, it was observed that the precipitate was uniformly dispersed as small, rounded particles throughout the structure. On the other hand, materials that flecked exhibited large masses of precipitate at the grain boundaries. Since size and distribution of the compound particles can be controlled by heat-treatment, this suggests that flecking can be eliminated by proper heat-treatment prior to exposure to high-temperature water.

Hydriding of zirconium during exposure to high-temperature water continues to receive major attention.

Samples of Zircaloy-2 and Zircaloy-4, with and without autoclave films, were exposed^{8,9} at 400°C to an atmosphere consisting of 93 vol.% helium, 4 vol.% hydrogen, and 3 vol.% CO. In 21 days the etched-but-unautoclaved samples of

both alloys were severely hydrided and very brittle. The autoclaved samples were found to contain 20- to 70-fold less hydrogen than the etched samples. However, the data for 53 days of exposure reveal that the hydriding rate is accelerating. This would indicate that the autoclave films offer only temporary protection against hydriding.

Investigations at Battelle¹⁰ have been concerned with the development of a high-strength corrosion-resistant zirconium alloy for cladding applications in a natural-uranium-fueled heavy-water-moderated power reactor. Room- and elevated-temperature hardness measurements were used to estimate the tensile strengths of the alloys, and corrosion behavior was evaluated by a 1000-hr static exposure to 300°C water. On the basis of a minimum hardness of 250 DPH and a maximum weight gain of 30 mg/dm² in corrosion tests, seven alloys containing 2 to 4 wt.% tin plus additions of molybdenum, iron, niobium, and/or nickel in the range 0.05 to 0.5 wt.% appear promising for the intended application. (W. K. Boyd)

Aluminum Corrosion

The solubility of aluminum corrosion products in contact with metallic aluminum in deionized water has been determined over the temperature range 80 to 350°C. Evidence has been developed at Hanford¹¹ which indicates that the oxide dissolution results in the formation of porous oxide films on aluminum exposed in refreshed dynamic systems. Dynamic corrosion-rate data were analyzed critically on the basis of a parabolic film growth and a linear oxide-degradation process acting simultaneously. The degradation rate was shown to be a function of the refreshment rate.

Investigations in France¹² have been concerned with the influence of titanium and zirconium additions on the structure of aluminum-iron-nickel alloys and the resultant effect on high-temperature-water corrosion behavior. It was found that, with prolonged heat-treatment, the eutectic structure of aluminum-iron-nickel alloy is subject to a coalescence. The addition of titanium and zirconium substantially retards corrosion both in cast and in wrought materials, but it does not affect the coalescence. The French were unable to explain the beneficial effect of the additions. Similar results have been obtained at Argonne and at Chalk River.

(W. K. Boyd)

Corrosion of Stainless Steels

In recently completed Bettis studies¹³ of the stress-corrosion cracking behavior of type 410 stainless steel, specimens representing different tempered conditions were stressed to 40,000, 60,000, and 80,000 psi in 300°F water. In general, stress-corrosion cracking was obtained only for those materials tempered at 650°F and stressed at 60,000 psi or above. No failures were observed for specimens tempered at 1150°F. Similar results have been observed previously at Knolls and Battelle.

Chromium plate was not found to be effective in eliminating cracking; in fact, in some instances the time to cracking was accelerated. Work at Knolls,¹⁴ on the other hand, has indicated that shotpeening, when properly incorporated into a product design, will minimize cracking of hardened type 410 stainless steel in high-purity water at temperatures up to 300°F.

Stainless steels and related iron-chromium-nickel alloys, because of their strength and resistance to oxidation, are being considered as cladding and barrier materials for gas-cooled reactors. Compatibility studies in various gases are being conducted by Oak Ridge¹⁵ and Aerojet.¹⁶ In nitrogen atmosphere, metals containing chromium are susceptible to nitriding. However, if small amounts of oxygen are present, an oxide scale is formed which retards or prevents nitriding. Although some cladding materials meet gas-compatibility requirements, they do not possess adequate strength. This suggests the use of a duplex system comprised of an outer layer having the necessary scaling resistance over a high-strength-alloy inner layer. Cladding materials that are being studied include iron-chromium-aluminum alloy, Inconel 702, Hastelloy R-235, Inconel X, Carpenter 20, and niobium. (W. K. Boyd)

Corrosion of Iron-, Nickel-, and

Cobalt-Base Alloys

In a study of the kinetics of oxidation of iron-30 to 78 wt.% nickel alloys, workers at General Electric¹⁷ confirmed the mechanism of oxidation in air at temperatures from 1100 to 1850°F as cation diffusion through the oxide scale. Anomalously high oxidation rates of the iron-78 wt.% nickel alloy (which also contained 3.8 wt.% molybdenum) near 1800°F were associated with auto-heating.

British workers¹⁸ investigated the effects of chromizing upon the elevated-temperature, air-oxidation resistance of various steels, nickel-base (Nimonic type) alloys, and cobalt-base (Jessop G-32) alloys. Their findings indicated that the nature of oxidation was altered in such a way as to decrease the oxidation rates substantially. The extent of selective grain-boundary oxidation was significantly diminished by the chromizing treatment.

Long-term corrosion tests of various high-temperature base-metal alloys in pressurized hot water or steam are being carried out at several laboratories. Autoclave tests by Martin Nuclear¹⁹ in 500°F water and steam (chlorinated, oxygenated, and with pH control) on Inconel resulted in very low corrosion rates (<1 mil/year) at the end of 500 hr, and no pits or cracks were found in stressed specimens in any of the media, confirming the observations of prior investigators. Hot water-steam loop tests on Croloy 16-1 and Inconel vessels are currently in progress at Martin.²⁰ Two Croloy components, removed after 950 and 2700 hr of test, showed excessive corrosion-product buildup at the location of the vapor-liquid interface. Inconel components, by comparison, showed very little attack.

Bettis workers¹³ have found very low corrosion rates on annealed or sensitized Inconel exposed for 1000 hr to 585°F ammoniated water or ammoniated or lithiated steam. Descaling problems have hampered the quantitative evaluation of results to some extent. These tests are continuing. In an additional study on the wear resistance of Inconel in reactor environments, it was found that Inconel-Inconel couples (threads, bushings, etc.) showed severe tendencies to gall. Chromium plating was effective in eliminating galling. None of the observed effects were associated with specific environments (lithiated or ammoniated water, etc.).

Continuing tests at Hanford⁹ have been in progress for two months. Data on the various alloys being investigated in deionized water at 680°F under 3200-psi pressure are shown in Table IV-1.

The French^{21,22} reported on the behavior of carbon, low-alloy, and stainless steels in CO₂ under pressure. The behavior of these steels was studied at temperatures from 662 to 1112°F, at pressures from 1 to 25 atm, for times to 6000 hr. The CO₂ atmosphere contained <0.1 per cent free oxygen. In general, the degree of

Table IV-1 CORROSION OF HIGH-TEMPERATURE ALLOYS⁹ IN PRESSURIZED DEIONIZED WATER AT 680°F

| Alloy | Two-month weight change, mg/dm ² |
|------------------------------|---|
| Fe-24 Cr-6 Al | +6.1 |
| Alfenol (Fe-16 Al) | +20 |
| Ferral (Fe-7 Cr-8.5 Al-2 Ti) | +20 |
| Modified Ferral | +8.4 |
| Hastelloys (various) | +11.6 to +24 |
| Inconel | +24.2 |
| Fe-30 Cr-1 Y | (Some weight loss, 1 month) |
| D-979 (Fe-45 Ni) | (Some weight loss, 1 month) |

oxidation was proportional to temperature, with rather high linear rates at 1112°F. Below about 930°F, oxidation was relatively slight. Increased pressures provided a beneficial effect, probably by promoting more compact and, hence, more protective scales. Chromium and aluminum were found to be beneficial alloying elements in enhancing oxidation resistance in CO₂, despite their propensity to oxidize selectively.

Several nickel-base alloys are being evaluated for fuel-pin cladding materials at Aerojet-General²³ by exposure in nitrogen at 1750°F and 200-psi pressure. The atmosphere contains 0.5 per cent oxygen and has a -30°F dewpoint. Protracted times are 10,000 hr. The reported information is shown in Table IV-2. Thermal

Table IV-2 WEIGHT CHANGE AND DEPTH OF INTERGRANULAR PENETRATION²³ OF NICKEL-BASE ALLOYS EXPOSED TO NITROGEN AT 1750°F

| Alloy | 1000-hr exposure | | 2500-hr exposure | |
|-----------------|------------------------------------|----------------------------------|-----------------------------------|----------------------------------|
| | Weight change,* mg/cm ² | Inter-granular penetration, mils | Weight change, mg/cm ² | Inter-granular penetration, mils |
| Inconel | +100 (9) | 0 | | |
| Inconel X | -5 (14) | 9 | | |
| Inconel 702 | +3.5 (15) | 3 | +8 | 0 |
| Hastelloy X | 0 (7) | 0 | +3 | 3 |
| Hastelloy R-235 | +6.5 (12) | 6 | +15 | Some |

*Parenthetical figures are parallel air-exposure values.

stability of mechanical properties was investigated briefly. After 1000 hr, hardening did occur in all alloys except Inconel and Hastelloy R-235, which were softer after exposure. After 2500 hr, Hastelloy X was moderately

strengthened; but it still retained good (20 per cent elongation) ductility.

Nickel, K-Monel, and niobium-1 wt.% zirconium alloy were exposed at Battelle²⁴ to flowing helium (some H₂O, N₂, O₂, CO₂, and CO impurities) for 250 hr at 960 and 1500°F under irradiation. Gettered (zirconium) and ungettered tori were exposed. Irradiation effects were not studied. Of the materials tested, only K-Monel in the ungettered container showed evidence of attack. This attack appeared as intergranular surface attack extending to a depth of about 2.5 mils after the 250-hr 1500°F exposure. The parallel niobium-1 wt.% zirconium sample showed no surface hardness increase. (E. S. Bartlett)

Corrosion by Liquid Metals

An Atomics International report²⁵ describes the salient features of the Large Components Test Loop (LCTL). This continuous flow system has been developed to meet the need for a test facility that can be used for corrosion and thermal-shock experiments of full-scale components anticipated for future sodium-cooled graphite-moderated reactors. The system is constructed primarily of 2 1/4 wt.% chromium-1 wt.% molybdenum alloy steel pipe and is capable of circulating continuously purified nitrogen-blanketed sodium at variable flow rates up to 1200 gal/min with temperatures up to 1000°F. Thermal-shock tests can be conducted with temperature transients as high as 500°F. Preliminary operational experience with the facility is being gained, and minor modifications are being made as a prelude to formal experimental work.

A report²⁶ originally issued in 1956 and recently declassified summarizes results from an investigation at Oak Ridge of the corrosion behavior of several high-temperature brazing alloys for use in sodium-air heat exchangers (fabricated of austenitic stainless steels or Inconel). The evaluations were carried out principally in a seesaw corrosion apparatus (usually, 1500°F for 100 hr). Alloys holding maximum promise included systems of nickel-silicon and boron, nickel-chromium-silicon-boron, and nickel-chromium-silicon. In general, precious-metal alloys were badly attacked by sodium, as were many silicon-free and chromium-free phosphorus-bearing materials. Alloys for sodium-fused salt radiator service were also studied. In this case the alloy systems nickel-chromium-phosphorus,

nickel-chromium-silicon-boron, nickel-silicon-boron, and nickel-germanium-chromium-silicon were especially favorable.

Attention is called to several bibliographies²⁷⁻²⁹ of various papers and articles that deal with liquid metals and liquid-metal systems. Report LM/TAB-13 is the most recent document in a regular series being issued by MSA Research Corporation.

Several organizations throughout the country are currently focusing attention on the potential for application of liquid-metal-vapor cycles in nuclear-powered space vehicles. One limitation being encountered in engineering studies along such lines is a lack of experimental physical-property data for the liquid and vapor states over the pressure and temperature ranges of interest. Fortunately, thermodynamic principles can be used to arrive at reasonable estimates of the more important properties. Such estimates are compiled in a document by Southwest Research Institute.³⁰ Fluids covered are liquid metals (mercury, cesium, rubidium, potassium, sodium, and lithium), liquid nonmetals (sulfur, phosphorus, and aluminum bromide), and gases (krypton, argon, helium, hydrogen, and aluminum chloride). Data presented include specific heats, enthalpies, entropies, densities, viscosities, vapor pressures, and heats of vaporization. Mollier charts (enthalpy-entropy) covering broad regions of interest are also presented. (J. H. Stang)

Radiation Effects

in Nonfuel Materials

Basic Studies

Irradiation hardening in crystalline solids deformable by a slip process is possible theoretically either by (1) dislocation source hardening or (2) lattice hardening, which increases the stress to move dislocations. In well-annealed metals, both types of hardening occur after neutron irradiation. However, in the case of LiF irradiated with 1-Mev electrons and tested in compression, lattice hardening governs the plastic deformation.³¹ The yield point, σ , varied with the dose, ϕ , according to the equation $\sigma = \sigma_0 + A[1 - \exp(-B\phi)]^{1/2}$; this relation was derived theoretically by considering dislocation movement through radiation-induced obstacles dispersed in the slip planes, plus trapping of point

defects on existing obstacles, which are probably clusters of point defects. The equation is intended to apply generally to crystals that deform by slip.

Defects in LiF generated by the $\text{Li}^6(n, \alpha)\text{H}^3$ reaction were studied by X-ray diffuse scattering and other X-ray diffraction methods.³² Results indicate defects with dimensions around 10 Å, increasing with both neutron dose and annealing temperature. Gas pressure due to fluorine and helium collection leads to development of cavities. Evidence was found for the formation of platelets of lithium in epitaxy with the LiF lattice. The X-ray measurements permitted following the formation and development of these imperfections when their atomic concentrations reached 10^{-4} .

Specpure monoclinic ZrO_2 subjected to fast-neutron irradiation did not transform to tetragonal, whereas baddeleyite did transform.³³ Commercial zirconia samples, originally partly in the metastable tetragonal form, did not change under neutron bombardment. Therefore, neutron irradiation cannot be considered equivalent to the process of heating to high temperature and quenching, and this appears to be evidence against the theory of phase transformation in zirconia by thermal spikes.

Measurements of increases in Young's modulus of copper following high-energy electron irradiation and subsequent recovery annealing have been interpreted in terms of dislocation pinning by various point defects.³⁴ Three recovery stages were noted and explained as: Stage IV at $+80^\circ\text{C}$, migration of single vacancies to dislocations; Stage III at -50°C , single interstitials, both stages occurring at somewhat lower temperatures than observed by resistivity measurement; and a recovery stage at -45°C , not observed by resistivity measurement, attributed to movement of interstitials lying close to dislocations and produced during irradiation by crowdion impact with the dislocation.

Marked radiation-enhanced diffusion was observed³⁵ in a water-quenched single crystal of iron-15 wt.% aluminum irradiated with 1.7×10^{18} neutrons/cm² and subsequently annealed. The ordering that occurred on annealing of the quenched sample was followed by measurement of the recovery of magnetic anisotropy, which, after a 1-hr anneal at 440°C , was nearly 10 times that of the unirradiated control.

Damage recovery is being studied at Hanford⁹ in a number of metals. Complete recovery of

the electrical resistivity of zirconium, irradiated to an integrated flux of 4.7×10^{18} nvt, occurred at 350°C . Nickel irradiated to 1.0×10^{19} nvt showed a drop in resistivity on annealing at 300°C . The lattice constant of copper decreased after an exposure of 9×10^{18} nvt, with no further change after 1.5×10^{20} nvt. X-ray line broadening decreased continuously with exposure. The X-ray-peak shifts with exposure agree with theoretical calculations for stacking-fault formation in copper. The lattice constant of iron increased up to 4.6×10^{18} nvt and then decreased with exposure to 1.5×10^{20} nvt, a behavior similar to that of molybdenum. A Russian paper³⁶ also reports marked X-ray line broadening in specimens of iron, chromium, nickel, and copper and in the alloys iron-nickel, iron-chromium, iron-manganese, and iron-tungsten exposed to 10^{20} nvt at $<80^\circ\text{C}$.

The density of lattice defects in copper foil produced by energetic alpha and lithium particles (from slow-neutron absorption in a layer of B^{10} on the foil surface) was many orders of magnitude greater than that in uncoated foil, as estimated from resistivity measurements.³⁷

A direct determination of equilibrium vacancy concentrations in high-purity aluminum was carried out by Simmons and Balluffi³⁸ by precise comparison of the linear dilatation $\Delta L/L$ and the fractional lattice-parameter change $\Delta a/a$ on the same sample at identical temperatures over the range 229 to 656°C . The values were equal to within about 1 part in 10^5 up to 415°C , but the values differed above this point according to the equation

$$3\left(\frac{\Delta L}{L} - \frac{\Delta a}{a}\right) = \exp(2.4) \exp(-0.76 \text{ eV}/KT)$$

At the melting point (660°C) the equilibrium concentration of defects, considered to be mainly single vacancies, is 9.4×10^{-4} . The divacancy concentration is estimated at less than 15 per cent, with a binding energy of ≤ 0.25 eV. The observed energy of formation, 0.75 eV, agrees with previous values obtained by quenching techniques and by high-temperature electrical-resistivity measurements³⁹ on the identical material. The first direct determination of formation entropy (2.0 to 2.4 eu), a value near that expected theoretically, was made. The method may be applicable to measurement of point defects in many materials.

Precision ultrasonic velocity and attenuation measurements were used by Truell⁴⁰ to determine the upper and lower size limits of a damaged region in a fast-neutron irradiated silicon single crystal. Assuming a spherical shape, the radius of the damaged region lies between 0.01 and 0.27 μ . From electrical-resistivity changes induced by bombarding copper, silver, and gold at 10°K with 1.5-Mev electrons,⁴¹ the threshold energy T_d for radiation-damage production in these metals is inferred to be 22, 30, and ≥ 40 ev, respectively. The fact that these values differ suggests a need to reconsider previous comparison of deuteron-bombardment experiment and theory, which indicated equality of T_d for the noble metals.

Helium injected into copper, aluminum, and beryllium, using these metals as targets for energetic alpha particles, appears on annealing as gas bubbles in regions where thermal vacancies originate.⁴²⁻⁴⁴ Grain boundaries appear to be the principal source of vacancies, provided their nature, misorientation, width, and direction are favorable. Line defects in the crystal are not vacancy sources, but they may be conductors. Vacancies produced during bombardment are mainly annihilated by the interstitial atoms also formed, and those remaining are too few to account for the helium bubbles observed. Similar formations of gas bubbles were reported in beryllium irradiated at elevated temperature⁴⁵ and in aluminum⁴⁶ and aluminum alloys,⁴⁷ where small grain size and the presence of second-phase particles enhanced bubble formation.

The rate of in-reactor damage accumulation in metal at a given temperature is the difference between the rate of damage production and the rate of recovery. Analysis by Gray⁴⁸ shows that, when interstitials and vacancies are produced in a constant neutron flux ϕ and recovery occurs by annihilation of interstitial-vacancy pairs, the rate of damage accumulation f_d at time t approaches the saturation value $f_{d(sat)} = \sqrt{c_1\phi/c_2}$, where c_1 and c_2 are the rate constants for production and recovery of damage, respectively. Thus, for a given material at constant temperature, the maximum number of displaced atoms will depend on $\phi = nv$, and a plot of f_d versus nvt will depend on the flux level.

Sosin⁴⁹ has prepared an excellent review of the present interpretation of radiation effects in metals. Another review article appears in the German literature.⁵⁰ A stochastically treated computer calculation of the cascade of displaced

atoms in metals induced by high-energy radiation has been carried out,⁵¹ assuming a screened coulomb interaction between atoms.

Baroody⁵² has considered theoretically the numbers of displaced atoms and vacant sites in the displacement cascade produced in compounds by the primary knock-on atoms on exposure to certain high-energy radiations. Assuming isotropic collisions involving pairs of free atoms, differential equations were developed with concentrations, collision cross sections, and mass ratios as parameters. Initial conditions defining threshold energies for displacement were introduced to account for binding of atoms.

Leibfried⁵³ has considered theoretically the encounter of long-range focusing collisions with stacking-fault areas of extended dislocations to produce Frenkel pairs, the number of which was calculated. The amount of enhancement of the radiation-damage rate by enhanced production of Frenkel pairs in heavily cold-worked metal is in qualitative agreement with the increased resistivity in pile-irradiated cold-worked copper. Furthermore, assuming that Frenkel-pair production in a dislocation line pins the dislocation, the increased number of pinning points per unit length of dislocation may be computed. The theoretical result on dislocation pinning by irradiation agrees within an order of magnitude with experimental data on copper.⁵⁴

(C. M. Schwartz)

Effects of Irradiation on Mechanical Properties

As indicated in Table IV-3, the strength of Inconel and Inconel X is usually increased and the ductility decreased by irradiation.⁵⁵ Property changes in the specimens irradiated at the higher temperatures were not as great, and the results indicate that saturation occurs after about 3.1×10^{19} fast neutrons/cm². A slight yield point was observed in the irradiated Inconel X, but the yield point was well developed in irradiated Inconel. Recovery of mechanical-property changes in annealed material irradiated at 50°C started at 200°C and was not complete at 600°C. Considerable recovery during irradiation at 250 and 300°C was suggested by the fact that the properties reached constant values near 7×10^{19} fast neutrons/cm². There was no evidence to suggest that radiation-enhanced aging occurred under any of the test conditions. There was no recovery of cold work in Inconel X after irradiation at 250°C.

Table IV-3 CHANGES IN ROOM-TEMPERATURE PROPERTIES OF NICKEL-CHROMIUM ALLOYS CAUSED BY NEUTRON IRRADIATIONS⁵⁵

| Condition | Integrated dose, 10 ¹⁹ neutrons(>500 ev)/cm ² | Irradiation temp., °C | Per cent change | | | Per cent change in elongation | |
|------------------|---|-----------------------|--------------------|----------------|---------------------------|-------------------------------|---------|
| | | | Proportional limit | Yield strength | Ultimate tensile strength | Total | Uniform |
| Inconel X | | | | | | | |
| Solution treated | 13 | 50 | +110 | +99 | +10 | -39 | -43 |
| Solution treated | 3.1 | 250 | +50 | +45 | +5 | -13 | -13 |
| Solution treated | 7.5 | 300 | +43 | +39 | +6 | -12 | -13 |
| Mill annealed | 13 | 50 | +176 | +133 | +20 | -27 | -33 |
| Mill annealed | 3.1 | 250 | +84 | +63 | +12 | +2 | 0 |
| Mill annealed | 7.5 | 300 | +67 | +48 | +14 | +11 | +9 |
| Cold drawn, 35% | 13 | 50 | +25 | +24 | +11 | -36 | -83 |
| Cold drawn, 35% | 3.1 | 250 | -4 | -4 | +6 | 0 | +9 |
| Inconel | | | | | | | |
| Cold drawn, 35% | 13 | 50 | +29 | +23 | +20 | -25 | -33 |
| Mill annealed | 7.5 | 300 | +50 | +45 | +9 | +8 | +10 |

The effects of irradiation on the room-temperature properties of type 304 stainless steel and 1100 aluminum after exposures on the order of 5×10^{20} neutrons(>0.1 Mev)/ cm^2 at 95°C have been studied.⁵⁶ The exposure caused the following percentage changes in the indicated properties:

| Material | Percentage changes in indicated properties | | | | |
|--------------------------|--|---------------------------|-------------------|-----------------|---------------|
| | Yield strength (0.2% offset) | Ultimate tensile strength | Reduction in area | Elastic modulus | Hardness |
| 1100 Al | +16 | +25 | | ~0 | +69 (R_H) |
| Type 304 stainless steel | +100 | +20 | -27 | +10 | +19 (R_A) |

The properties of the aluminum changed continuously as a function of dose, whereas those of stainless steel leveled out above 4×10^{20} fast neutrons/ cm^2 . Appreciable stress relief occurred in elastically stressed stainless steel but not in the aluminum.

A program¹² has been initiated to establish the nature of changes in mechanical properties of Zircaloy-2 during irradiation. Two specimens of annealed material are being evaluated (one in an in-reactor capsule and the other in the laboratory) to provide direct comparison. No creep was observed up to 30,000 psi at 260°C , when both specimens began to exhibit creep. The creep rate of the in-reactor specimen ap-

peared greater than that of the ex-reactor specimen. More accurate data are being obtained.

(B. C. Allen)

Selected Metallurgical Aspects of Cladding and Structural Materials

Niobium

Union Carbide⁵⁷ reports the existence of a NbAl compound that has tentatively been identified as a sigma phase having an aluminum content greater than Nb_3Al . X-ray diffraction data confirm this phase and place its composition at approximately 34 at.% aluminum.

(J. A. DeMastry)

Iron-Base Alloys

Russian⁵⁸ investigators have studied the influence of annealing time at 1200°C on the structure of iron-chromium-aluminum alloys. An increasing number of solid, brittle, and non-magnetic inclusions of a new phase appear with increased annealing time. The resistance to oxidation drops with increase in holding time. It is thought that these results are caused by decomposition occurring during annealing, thus bringing the alloys to an approximate state of equilibrium.

(J. A. DeMastry)

Yttrium

Nuclear Metals workers report⁵⁹ a study of the fabrication of yttrium metal. They state that

yttrium is fully recrystallized between 1000 and 1100°F with several varying amounts of cold work. Annealing at 1100°F appeared to be satisfactory for stress-relieving material that was cold worked 25 per cent. (J. A. DeMastry)

Diffusion Studies

A study of the diffusion of uranium with several transition metals was reported by the French.⁶⁰ Electron-probe microanalysis was used to obtain concentration-versus-penetration curves for the systems of uranium with molybdenum, molybdenum-uranium, niobium, niobium-uranium, zirconium, and titanium. From these curves, it is possible to determine (1) the diffusion coefficients and the activation energies as functions of the concentration and (2) the equilibrium diagram in multiphase regions. The growth kinetics of the various zones was also studied, and the corresponding activation energies were calculated. The Kirkendall effect was investigated, and the intrinsic coefficients were calculated.

In studies conducted by Hanford,⁶¹ an expression has been developed for the penetration of uranium, by diffusion through a nickel barrier, into an aluminum alloy at 450°C. The expression, which applies when the time is greater than 40 days, is $x^2 = 0.335 (t - 20)$, where x is the penetration of uranium in thousandths of an inch and t is the time in days. Solid-uranium diffusion in AlSi-bonded Hanford fuel elements has also been studied, and results are presented for penetration at 300°C.⁶¹

In an investigation of the thermal diffusion of hydrogen in Zircaloy-2 at Bettis,⁶² it was shown that substantial hydrogen migration to the cold side occurred, thus causing the precipitation of a large amount of metal-free zirconium hydride there. This confirms postulated behavior.

The diffusivity of helium through aluminum has been measured in the temperature range 400 to 550°C by two different techniques at Materials Research Corporation.⁶³ In the first technique an aluminum sheet cathode was charged with gas by bombarding it with helium ions accelerated by a potential, and, subsequently, evolution from a diffusion sandwich of this material was measured. The diffusivities obtained exhibited the expected temperature dependency. In the second technique the gas was introduced into an aluminum-base lithium alloy by neutron irradiation and subsequently evolved;

however, the results did not vary with temperature in the expected fashion.

(D. C. Carmichael)

Selected Mechanical Properties of Cladding and Structural Materials

Zirconium Alloys

Attempts to design nuclear reactors utilizing Zircaloy-2 having some degree of cold work have prompted studies at Battelle^{5,64} and Hanford^{6,65} on the properties of 15 to 45 per cent cold-worked Zircaloy-2 at elevated temperatures. Battelle has been investigating the creep properties of 15 per cent cold-worked Zircaloy at 150 to 400°C under constant- and cyclic-temperature conditions. Some of these tests are long term, 10,000 to 20,000 hr. Hanford workers have been examining the creep properties as affected by cold work within the 15 to 45 per cent range. They found that, for test temperatures up to 400°C, residual cold work improves the creep strength of Zircaloy-2. The initial deformation upon application of load is reduced, and subsequent creep rates are reduced. However, higher creep rates were observed for 45 per cent cold-worked material than for 25 per cent cold-worked material. Second-stage or steady-state creep was not obtained until after 2000 hr. The tensile properties of Zircaloy-2 also have been measured by Atomics International⁶⁶ at elevated temperatures, and creep data have been developed for moderate loads at 565°C. Because the state of stress in many cases alters the tensile properties of a material, workers at Knolls⁶⁷ have been investigating the effect of combined stresses on the yield and fracture behavior of Zircaloy-2. They noted that the fracture ductility decreased with increasing biaxiality; the minimum was found at balanced biaxial tension where the room-temperature fracture ductility, expressed as effective strain, was 29 per cent.

Zircaloy-4, which is nominally the same composition as Zircaloy-2 with the nickel removed, has been tested in creep at 350°C by workers at Bettis.¹³ It was tested in two heat-treated conditions—as received and base annealed. As-received material was mill annealed for 15 min at 843°C, and the base annealed material was annealed for 20 hr at 750°C in vacuum and furnace cooled. Table IV-4 gives the minimum

Table IV-4 CREEP RATE¹³ OF ZIRCALOY-4 AT 350°C

| Stress, psi | Minimum creep rate, hr ⁻¹ |
|----------------------|---|
| <i>As Received</i> | |
| 25,000 | 2.65×10^{-5} |
| 28,000 | 4.20×10^{-4} |
| <i>Base Annealed</i> | |
| 24,000 | 1.80×10^{-5} |
| 25,200 | 1.59×10^{-4} |
| 27,000 | 9.0×10^{-4} |
| 29,200* | 4.7×10^{-1} |

*Ultimate tensile strength.

creep rates for several stresses which are similar to those obtained for Zircaloy-2.

The creep rate for a zirconium-17.5 wt.% niobium alloy at 500°C has also been determined. The material was homogenized at 1000°C for 24 hr, was tested at 33,100 psi at 500°C, and had a minimum creep rate of 1.5×10^{-4} in. per inch per hour.

Other zirconium-base alloys are being studied at Knolls.⁶⁸

Attempts were made by Battelle¹⁰ researchers to extend the useful service temperature range of zirconium alloys by additional alloying to increase the strength and corrosion resistance. Approximately 100 ternary and quaternary sponge-zirconium alloys were screened. Alloy additions studied included 2 to 4 wt.% tin, 0.5 to 2 wt.% molybdenum, and 1 to 3 wt.% niobium. Room- and elevated-temperature hardness measurements were used to estimate the tensile strengths of the alloys, and corrosion resistance was evaluated by 1000-hr exposures to static 300°C water. The following alloys appeared to be promising for higher temperature application:

Zirconium-2.0 wt.% tin-0.5 wt.% molybdenum

Zirconium-2.0 wt.% tin-2.0 wt.% niobium-0.1 wt.% iron-0.05 wt.% nickel

Zirconium-2.0 wt.% tin-3.0 wt.% niobium-0.1 wt.% iron-0.05 wt.% nickel

Zirconium-3.0 wt.% tin-0.1 wt.% iron-0.05 wt.% nickel

Zirconium-3.0 wt.% tin-0.5 wt.% molybdenum-0.1 wt.% iron-0.05 wt.% nickel

Zirconium-3.0 wt.% tin-0.5 wt.% molybdenum-1 wt.% niobium-0.1 wt.% iron-0.05 wt.% nickel

Zirconium-4.0 wt.% tin-0.5 wt.% molybdenum

Stainless Steel

The use of stainless steel as structural and cladding materials in nuclear reactors is limited by the reactor concept and its application and operating temperatures. The thermal-fatigue properties of AISI type 347 stainless steel have been given special consideration by workers at Knolls.⁷⁰ Expected application is for an in-pile tube in the ETR. Scaled-down sections of an in-pile tube were installed in the HF-2 loop and subjected to a total of 10,020 thermal cycles and 8 pressure cycles. Maximum tangential stresses observed were 16,520 psi from pressure and 98,680 psi from internal heat generated. There was no evidence of growth or cracking resulting from the test outlined. Other studies made on stainless steel by U. S. Steel Corporation⁷¹ have indicated that small variations of combined amounts of carbon, nitrogen, and phosphorus can have a marked effect on the 100- and 1000-hr creep-rupture strengths of type 316 stainless steel. Variation of properties of 14 different 18 wt.% chromium-8 wt.% nickel-molybdenum steels at 1100, 1300, and 1500°F could be accounted for by variation in chemical factors in about half of the specimens. There are additional variations related to differences in thermal and mechanical history. Size was also observed to influence variations in properties. The high-temperature strengths of some iron-chromium-aluminum alloys were evaluated by the Russians⁷² at 600 and 1200°C under various bending stresses.

Battelle⁷³ researchers have investigated the effect of yttrium additions on the fabrication and tensile properties of two modified stainless steels. These alloy compositions were nominally (1) 55 wt.% iron-21 wt.% nickel-17 wt.% chromium-2.5 wt.% molybdenum-1.0 wt.% niobium-0.03 wt.% carbon-0.5 wt.% manganese-0.5 wt.% silicon with additions up to 1.5 wt.% yttrium and (2) 36 wt.% iron-37 wt.% nickel-18 wt.% chromium-2.5 wt.% molybdenum-1.5 wt.% niobium-1.0 wt.% aluminum-0.05 wt.% carbon-0.5 wt.% manganese-0.5 wt.% silicon with up to 2.0 wt.% yttrium additions. At temperatures up to 1850°F, yttrium had virtually no effect on the yield and the ultimate strengths. Between 1000 and 1750°F the

ductility of fabricable alloys was increased with increasing additions of yttrium.

Niobium

A study was conducted by Battelle⁷⁴ workers to improve the elevated-temperature strength of niobium by alloying. Greatly improved strengths were achieved at 1200 and 1470° F, and improved fabrication techniques were found. The 0.2 per cent offset yield strengths of a cold-worked niobium-1.84 wt.% chromium alloy at 1200 and 1470° F were 107,000 and 69,000 psi, respectively, and the niobium-4.33 wt.% zirconium alloy had a 0.2 per cent offset yield strength of 69,000 psi at 1470° F. The strengthening of pure niobium by solid-solution alloying with up to 10 wt.% titanium, molybdenum, or tungsten has been investigated at Du Pont.⁷⁵ The strengthening effects at 2000° F were determined by creep-rupture, hot-tensile, and hot-hardness testing. Recrystallization studies were also made. All three solute elements were found to improve the high-temperature strength of niobium. Tungsten was the most effective, and the niobium-titanium alloy showed maximum strengthening at about 9 at.% titanium. The recrystallization temperature was raised with increasing additions of molybdenum and tungsten, but the temperature was not changed appreciably by increasing the titanium content.

A series of niobium-base alloys was prepared from powder, in both the attritioned and dehydrided conditions, for Bettis.¹³ Methods of preparation were essentially to cold press, jacket and hot forge to full density, and anneal for 12 hr. The tensile properties at room and elevated temperatures, as determined at Bettis, are given in Table IV-5.

Miscellaneous Metals and Alloys

Other refractory metals, such as tungsten, have applications for extreme temperatures and fast-neutron reactors. The short-time creep-rupture properties of tungsten have been determined by Los Alamos⁷⁶ at 2250 to 2800° C. Tungsten retains an appreciable creep strength and reasonable ductility in the temperature range 2250 to 2800° C. The mode of creep failure is predominantly intercrystalline. Tungsten sustained stresses up to 7000 psi for times up to 4 hr. The creep behavior of molybdenum was studied by the same researchers⁷⁷ on commer-

cial powder-metallurgy rod at 1000 to 2500° C for stresses up to 9000 psi.

Among the lighter metals of interest for nuclear-reactor application are aluminum and magnesium. Atomics International⁷⁸ has investigated several aluminum powder-metallurgy products because of their good elevated-temperature properties. By tensile and creep studies, Atomics International has confirmed the superiority of APM (Aluminum Powder Metallurgy) alloys over conventional wrought aluminum alloys in the temperature range 800 to 1000° F. The increased elevated-temperature strength is apparently due to the dispersion strengthening effect of small oxide particles. Results from notched tensile specimens indicated that APM products are not notch sensitive for stress concentration factors of 2.5 and 5.0. The creep strength of these products at 900° F is somewhat lower than that reported by commercial suppliers.

Other light-metal alloys, magnesium-0.75 wt.% cerium-0.5 wt.% zinc-0.5 wt.% zirconium and magnesium-6.5 wt.% aluminum-1.0 wt.% zinc, have been investigated by the Canadians.⁷⁹ The creep properties were determined at 50 and 80° C at various stresses. Specimens were prepared from longitudinal and transverse sections of an extruded tube of the first alloy. The extrapolated creep rate of 1×10^{-5} per cent per hour was attained at 50 and 80° C by 12,500 and 9500 psi, respectively. This alloy showed less creep strength in the transverse direction. The ultimate strengths at 50 and 80° C were 28,000 and 25,000 psi, respectively. Creep tests performed on the magnesium-6.5 wt.% aluminum-1.0 wt.% zinc alloy at 50° C revealed creep strengths similar to those for the magnesium-6.5 wt.% aluminum-1.0 wt.% zinc-0.25 wt.% manganese alloy.

Recent Literature Surveys

Several comprehensive literature surveys have been made for some structural materials. A review of the qualifications of Zircaloy-2 and zirconium as reactor-core structural materials has been compiled at Hanford⁸⁰ for the purpose of reporting changes in problems of application and laboratory data during the past five years. It contains 85 references. The Defense Metals Information Center published⁸¹ a review of the properties of refractory materials, using an

Table IV-5 TENSILE PROPERTIES¹³ AT ROOM TEMPERATURE AND AT 500 AND 900°C

| Heat-treatment, °C | Alloy composition (balance Nb), wt. % | Type | Test temp.,* °C | Yield strength (0.2% offset), 1000 psi | Tensile strength, 1000 psi | Elongation, % | Density, % |
|-----------------------|---|-------------|-----------------------|--|----------------------------------|------------------|---------------|
| 1300 | 10 Zr | Hydride | RT | 65.5 | 81.1 | 22 | 99.1 |
| 1260 | 15 Zr | Hydride | RT | 83.1 | 97.9 | 17 | 95.5 |
| 1260 | 15 Zr | Hydride | 500 | 60.7 | 83.3 | 18 | |
| 1260 | 15 Zr | Hydride | 900 | 41.0 | 81.9 | 16 | |
| 1260 | 15 Zr | Attritioned | RT | 86.9 | 89.7 | 22 | 98.5 |
| 1260 | 15 Zr | Attritioned | 500 | 58.3 | 80.6 | 8 | |
| 1260 | 15 Zr | Attritioned | 900 | 54.2 | 72.0 | 6 | |
| 1050 | 50 Zr | Hydride | RT | 121.1 | 127.8 | 2 | 99.0 |
| 1050 | 50 Zr | Hydride | 900 | 92.9 | 99.4 | 2 | |
| 1050 | 50 Zr | Attritioned | RT | 117.8 | 122.6 | 2 | 97.3 |
| 975 | 25 Zr | Hydride | RT | 96.4 | 101.1 | 3.5 | 100 |
| 975 | 25 Zr | Hydride | 500 | 79.3 | 93.1 | 8.5 | |
| 975 | 25 Zr | Hydride | 900 | 73.3 | 83.9 | 9.0 | |
| 1390 | 5 Mo | Attritioned | RT | 45.7 | 61.8 | 9.5 | 97.6 |
| 1390 | 5 Mo | Attritioned | 500 | 28.7 | 48.8 | 13.5 | |
| 1390 | 5 Mo | Attritioned | 900 | 27.5 | 50.3 | 7.5 | |
| 1340 | 5 Mo | Hydride | RT | 53.0 | 71.8 | 30 | 99 |
| 1340 | 5 Mo | Hydride | 500 | 35.2 | 63.7 | 20.5 | |
| 1340 | 5 Mo | Hydride | 900 | 42.7 | 68.5 | 17 | |
| 1340 | 10 Mo | Attritioned | RT | 56.6 | 79.1 | 21.5 | 99.1 |
| 1340 | 10 Mo | Attritioned | 500 | 43.5 | 62.4 | 15.5 | |
| 1340 | 10 Mo | Attritioned | 900 | 40.3 | 68.0 | 15.0 | |
| 1340 | 10 Mo | Hydride | RT | 60.2 | 82.7 | 26.5 | 99 |
| 1340 | 10 Mo | Hydride | 500 | 44.3 | 69.7 | 18.5 | |
| 1340 | 10 Mo | Hydride | 900 | 41.4 | 69.1 | 16 | |
| 1340 | 3 V-5 Zr | Attritioned | RT | 67.1 | 86.4 | 10.5 | 97.9 |
| 1340 | 3 V-5 Zr | Attritioned | 500 | 49.3 | 75.3 | 13 | |
| 1340 | 3 V-5 Zr | Hydride | RT | 65.7 | 86.7 | 29 | 99.3 |
| 1340 | 3 V-5 Zr | Hydride | 500 | 49.8 | 74.3 | 21 | |
| 1340 | 3 V-5 Zr | Hydride | 900 | 47.8 | 79.9 | 14 | |
| 1340 | 5 Mo-5 Zr | Attritioned | RT | 64.5 | 81.9 | 20 | 98.6 |
| 1340 | 5 Mo-5 Zr | Attritioned | 500 | 55.6 | 61.4 | 23 | |
| 1340 | 5 Mo-5 Zr | Hydride | RT | 61.5 | 77.5 | 25 | 97.6 |
| 1340 | 5 Mo-5 Zr | Hydride | 500 | 47.1 | 65.3 | 24 | |
| 1340 | 5 Mo-5 Zr | Hydride | 900 | 38.4 | 68.3 | 21.5 | |
| 1340 | 2.8 V | Attritioned | RT | 41.9 | 62.2 | 25 | 99.1 |
| 1340 | 2.8 V | Attritioned | 500 | 30.9 | 51.4 | 23 | |
| 1340 | 2.8 V | Hydride | RT | 40.5 | 64.7 | 29.3 | 99.5 |
| 1340 | 2.8 V | Hydride | 500 | 29.7 | 52.1 | 28 | |
| 1340 | 2.8 V | Hydride | 900 | 28.3 | 52.3 | 27 | |
| 1315 | 5.8 V | Attritioned | RT | 66.3 | 88.5 | 25.4 | |
| 1315 | 5.8 V | Attritioned | 900 | 40.0 | 74.5 | 17.0 | |
| 1315 | 5.8 V | Hydride | RT | 51.1 | 88.2 | 27.5 | |
| 1315 | 5.8 V | Hydride | 500 | 48.1 | 73.2 | 21.5 | |
| 1315 | 5.8 V | Hydride | 900 | 42.3 | 72.4 | 23.5 | |

*RT = room temperature.

1800°C melting point as the cutoff to differentiate between refractory and nonrefractory materials. This survey covers properties of 122 refractory materials, including graphite, 12 refractory metals, 18 carbides, 13 oxides, 12 nitrides, 17 silicides, 19 borides, 22 sulfides, and 8 inter-metallic compounds. Another recent DMIC report⁸² deals with the physical and mechanical properties of the niobium and niobium-base alloys. (F. R. Shober)

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Melting, Casting, Heat-Treatment, and Hot Working

Interest in the casting of refractory materials is evident from reports from several sources. The Bureau of Mines¹ reports on the static and centrifugal casting of molybdenum. Using consumable-electrode skull type arc-melting techniques, metal yields as high as 70 per cent of the electrode weights were obtained. Although some difficulties (surface condition and internal soundness) were experienced with static graphite molds, the properties of the resulting metal were similar to those found in unworked shapes prepared by powder-metallurgy techniques.

The Defense Metals Information Center² recently published the text of a talk that, in part, briefly summarized the melting and fabrication of a number of refractory materials and concluded that significant developments will be forthcoming. Notable among the anticipated developments were electron-beam melting, purifying, and consolidating.

Yttrium has been the subject of several fabrication studies. The Bureau of Mines, Nuclear Metals, and General Electric have all participated in research on this topic.³⁻⁶ Bohlander, Williams, and Huffine of General Electric summarized and reported on experiences with this metal at the annual ASM meeting held in Chicago Nov. 3 to 5, 1959. The most serious difficulty experienced in the fabrication of yttrium was found to be its sensitivity to oxygen-impurity content. Yttrium containing 500 ppm oxygen is considered ductile since it can be cold reduced 65 to 90 per cent without cracking; yttrium containing 1000 to 3000 ppm can be worked successfully only at 1650°F. Hot working is complicated by the rapid oxidation that occurs above 1400°F, requiring protection by a special atmosphere or cladding. The recrystallization temperature was reported to be between 1000 and 1200°F. Because of its reactive nature, yttrium must be machined with caution. Sharp

tools with a positive rake angle, high-flash-point lubricants and oils for machining, and chip storage were recommended.

A patent recently issued to Smith⁷ describes a method for rolling uranium under nonoxidizing conditions at relatively low temperatures (200°C). Reduction steps of approximately 15 per cent per pass to a total of 90 per cent without edge cracking are claimed if the uranium is heated in oil and rolled after allowing the oil to drain so that only a thin film remains. No appreciable increase in embrittlement is said to occur.

In the development of high-strength niobium alloys for use at elevated temperatures, Battelle employed a fabrication technique that consisted of warm forging and rolling the alloy at 1000°F in air.⁸ This technique was effective in overcoming the cast structure of the arc-melted alloy ingots. Following the warm reduction, it was possible to cold roll to 30-mil sheet with only minor edge cracking.

The presence of stringers in zirconium alloys, particularly Zircaloy-2, has been causing concern for some time. A patent recently issued to Pickleseimer⁹ claims, in part, that the fabrication and heat-treatment methods cited will prevent stringer formation and, in addition, will provide a randomly oriented crystal structure. The method consists of working at a temperature outside the alpha-plus-beta range (810 to 970°C), holding at a temperature above 970°C for 30 min, cooling rapidly to room temperature, reducing at least 20 per cent by working, and then annealing at a temperature of 700 to 810°C.
(E. L. Foster)

Cladding

Cladding by Rolling and Swaging

Although some additional work is in process on the study of roll cladding dispersion type

fuels, the major emphasis is currently on the cladding of UO_2 by swaging.

Over the past several years, Babcock and Wilcox¹⁰ has been developing a process for the manufacture of UO_2 fuel elements by use of the rotary-swaging technique. This process is intended for the fabrication of slightly enriched fuel rods of stainless-steel-clad UO_2 for the N.S. Savannah. The swaging technique was selected as the reference process for the preparation of experimental rods since higher production rates had been experienced with this method than with pelletizing. The latter process, however, is at present still the reference process for fabrication of the actual power-reactor core.

An investigation has also been initiated at Oak Ridge¹¹ to evaluate rotary swaging as a method for preparing stainless-steel-clad UO_2 fuel rods. In this study, it has been found that fused and ground UO_2 cold swages to the highest density, 91 per cent of theoretical, at a reduction in area of approximately 55 per cent. Ceramic grades of the oxide were swaged to a density of 75 per cent and rod milled and high fired to 85 per cent of the theoretical density. The swageable grades of the oxide are characterized by high-tapped bulk density and low surface area. Hot swaging at 800°C produced improved densities; however, variations in wall thickness are increased by swaging at temperature or by cold swaging in excess of 70 per cent reduction. Bonding has been observed, however, between particles during swaging at 800°C.

As-swaged densities as a function of temperature have been measured at Oak Ridge¹² from room temperature to 1000°C. The optimum densities are obtained at 800°C with an appreciable dropoff in densities with increased swaging temperatures above 800°C.

The feasibility of producing annular, internally and externally clad fuel tubes by swaging is also being investigated.¹² Work to date has been concerned mainly with swaging without using a mandrel to control inner-sheath geometry. Specimens prepared in this manner have been swaged to densities of about 7 per cent less than that obtained with a solid rod swaged to a similar reduction.

The fabrication of a metal fiber- UO_2 fuel element has been demonstrated by Martin¹³ to be feasible with rod type fuel elements containing

up to 20 vol.% fiber. Cladding materials consisting of zirconium, Zircaloy-2, type 304 stainless steel, and type 6061 aluminum alloy were employed in a study aimed at obtaining high-density UO_2 by swaging. Zirconium, Zircaloy-2, and type 304 stainless steel, when used as cladding materials, produced equivalent as-swaged densification of the oxide. However, the use of type 6061 aluminum alloy resulted in a decrease of approximately 2 per cent in the as-swaged density. The wall thicknesses of the different cladding materials had no effect on the final swaged density. The stainless-steel cladding material showed only a slight roughening, the aluminum alloy cladding exhibited a much greater amount of roughening, and the zirconium and Zircaloy-2 cladding materials exhibited severe crack formation as a result of swaging.

Graphite, molybdenum, and niobium appeared to offer the most promise as fiber materials; however, tests with graphite proved unsatisfactory. Molybdenum as a fiber material has been emphasized in studies to date. Specimens containing fibers of molybdenum swaged to a reduction of 40 per cent exhibited reasonably good radial fiber orientation. Increased reductions on specimens of relatively large diameters produced less favorable fiber orientations.

In preparing large numbers of swaged UO_2 fuel elements, Hanford¹⁴ was able to achieve constantly improving yields (approaching 90 per cent over a one-month fabrication period). Swaging experiments were performed with Zircaloy-2-clad oxide fuel elements. It was found that there is an optimum swaging reduction that produces a maximum swaged density. Swaging beyond this point results in decreased densities. It is felt that work hardening of the Zircaloy-2 cladding is responsible for this behavior.

It has been reported¹⁵ that Zircaloy-clad UO_2 fuels for possible use in the NPD after the initial charge are being prepared by rotary swaging. Previously, it was reported that Zircaloy-clad fuel rods in excess of 1 in. in diameter prepared by this technique contained severe defects. The defects have been avoided by the use of adjustable anvil blocks on the swager to give a shorter stroke to the swaging blow.

Although gross splitting of Zircaloy sheaths swaged cold to reductions of 35 to 50 per cent

has been avoided by warm swaging at 600°F, the presence of cracks along the inside surface of the wall has been noted in some specimens. Zircaloy sheaths on swaged oxide fuel elements 0.6 in. in diameter or larger still showed some defects after swaging to a 45 per cent reduction in area at 500°C. Intermediate anneals will be studied in an attempt to minimize these defects.

Cold-swaged densities of 92 per cent were achieved at Savannah River¹⁵ with Zircaloy-2 and stainless-steel sheaths 0.022 in. in thickness. Cladding materials of mild steel and aluminum were also studied. Maximum swaged densities for the fused oxide in each of these cladding materials were as follows: 92 to 93 per cent with type 316 stainless steel for a wall thickness of 0.032 in.; 92 per cent with Zircaloy-2 for a wall thickness of 0.022 in.; 88 to 89 per cent with mild steel for a wall thickness of 0.065 in.; and approximately 86 to 87 per cent with aluminum for a wall thickness of 0.035 in.

Experiments in which fused oxide was swaged in stainless steel varying in wall thicknesses from 0.065 in. to as low as 0.007 in. showed that densities as high as 92 per cent of the theoretical density could be obtained.

Also, flat rolling of UO₂ in stainless steel followed by mechanical twisting is being investigated¹⁵ as a method of fabricating a twisted-ribbon fuel element. Densities as high as 86 per cent of the theoretical density were obtained by rolling. Twisting the rolled ribbons 180° per foot lowered the densities, however. The rectangular shape of the ribbon was believed to have produced less favorable densities with this type of element. Some evidence of laminar cracking was also observed in the cores, which was attributed to a combination of rolling stresses and sheath springback.

Combustion Engineering and Aero projects¹⁶ are conducting a joint program to apply the principles of ultrasonic welding to the cladding of nuclear fuel plates. The initial studies will ascertain the feasibility and economics of the process. Power levels will be studied for cladding-to-cladding and cladding-to-core bonds and the achievement of edge and end bonds in picture-frame fuel elements. Aluminum and stainless-steel cladding materials will be employed in the initial studies. Normally, UO₂ dispersion fuels will be employed as core materials.

(E. S. Hodge)

Diffusion Bonding

Attempts to use copper-eutectic diffusion bonding¹⁷ of oxide plate fuel elements for the PWR Core 2 have been discontinued in favor of gas-pressure bonding. These bonding processes have been described in earlier issues of *Reactor Core Materials*. The deciding factor was the adverse effects of the copper utilized in eutectic diffusion bonding on the corrosion resistance of the Zircaloy cladding. It was found, for example, that copper diffusion to the surface of the cladding during bonding (at 1800 to 1900°F) resulted in a surface copper concentration of about 0.4 wt.% when 15-mil-thick cladding was used and 0.1 wt.% when 20-mil-thick cladding was used. At the high copper level, corrosion of the Zircaloy is accelerated, and high hydrogen pickup results. In compartments that were intentionally caused to be defective to expose bonds, where copper concentration is a maximum (about 1 wt.%), this effect is exaggerated in long-time tests in 680°F water. Since no foreign elements are involved in the gas-pressure-bonding process, it has supplanted copper-eutectic diffusion bonding as the reference process for concentrated development effort applicable to the manufacture of PWR Core 2 fuel elements. Some work^{17,18} has been continued, however, on the use of multiple bonding agents such as iron-copper to minimize the adverse corrosion effect of copper in the eutectic-diffusion-bonding process. Significant improvement has been indicated thus far in corrosion tests.

Methods were investigated at Sylvania¹⁹ for the fabrication of composite sheets consisting of type 310 stainless steel bonded to one side of either molybdenum or niobium sheet, utilizing a barrier material between the two sheets to minimize diffusion. Composites were fabricated by hot pressing at 1050°C for 35 to 45 min under a pressure of 2000 psi, followed by hot rolling. On the basis of tensile strength, thermal-shock resistance, minimum diffusion during long-time temperature tests, and minimum formation of intermetallic compounds in the bonds, the best barrier materials found were nickel for molybdenum-stainless steel composites and iron for niobium-stainless steel composites. Butt welds made on the composite sheets resulted in stresses and distortion in the weldments due to differences in the thermal coefficients of expansion between the stainless steel and the refractory metals.

In conjunction with Savannah River,^{20,21} a program on fabrication development and testing of metallurgically bonded joints between Zircaloy and stainless-steel tubes is being conducted. Only preliminary results have been reported thus far.

A U. S. patent has been issued on a process developed at Battelle.²² This process involves bonding of uranium to iron, cobalt, nickel, or their alloys. The bonding is accomplished by first electrodepositing silver to surfaces, amalgamating or alloying the silver layer with mercury or indium, and then pressure bonding and heating to complete bonding.

(D. C. Carmichael)

Coextrusion

The feasibility of coextruding stainless-steel-clad uranium elements was investigated by Hanford.¹⁴ Three stainless-steel-clad uranium billets were canned in copper and preheated to 640°C just prior to extruding; however, this treatment was unsuccessful. The stainless-steel cladding broke apart into approximately 1-in.-long rings around the uranium core during extrusion. An increase of 25 per cent in extrusion force was required over a similar Zircaloy-2-clad billet.

Investigations are being conducted by Nuclear Metals²³ on the coextrusion of uranium-10 wt.% molybdenum alloy with beryllium. Several rods have been extruded in an attempt to obtain integral end seals. The rod dimensions were on the order of 0.200 in. in diameter by 5 ft long with a cladding thickness of 0.010 to 0.020 in. The extrusion reduction was 25:1, and the extrusion temperature was 1950°F. Fairly uniform seals were obtained where there was good stiffness compatibility between core and cladding. Future work will be done to obtain a core alloy to give a better stiffness compatibility.

Hanford²⁴ has fabricated fuel-element rods 0.950 in. in diameter by 5 ft long containing a fuel material of aluminum-7.35 wt.% plutonium alloy clad with aluminum (X-8001 alloy). The cladding thickness was 0.040 to 0.120 in. with a core length of 55 to 57 in.

A program to evaluate current coextrusion procedures for the extrusion of Zircaloy and zirconium-clad uranium rods was conducted by Bridgeport Brass.²⁵ From the limited amount of work completed, it was concluded that the

coextrusion techniques used at present need only refinement to make the process an efficient one.

(C. B. Boyer)

Extrusion Cladding

Tubes. The technical feasibility of extrusion cladding the inside and outside of nickel-plated uranium tubes with a 30-mil aluminum layer has been demonstrated.²⁶ Tubes about 3.4 in. in outside diameter by about 2.9 in. in inside diameter and up to 4 ft long were clad.

Bond strengths on the 3.4-in.-OD tubes ranged from 3000 to 12,000 psi. Of the 16 tubes tested, 10 had bond strengths averaging 5000 psi and were adjudged to be adequate for reactor service, and six had a high percentage of nonbonded areas. It was found that none of the bonds achieved the 27,000-psi maximum which had been observed previously on tubes clad on the inside only. Also, the bond strength on the outside generally exceeded that on the inside by 3000 psi.

In the extrusion cladding of tubes, ultrasonic energy has been utilized in order to increase the extrusion speed at a given ram load. The application of 1800 to 2300 watts and 20,000 cps to certain tools increased the extrusion speed by 100 per cent when cladding the aforementioned 3.4-in.-OD tubes.²⁶ Other experiments²⁷ showed that, when cladding 2.75-in.-diameter steel tubes at a constant force of 40 tons, the extrusion speed could be increased up to 250 per cent by the application of ultrasonic vibrations. The improvement in extrusion speed appeared to be attributable to a reduction of both die friction and container friction under the influence of ultrasonic energy. (H. J. Wagner)

Nonelectrolytic Chemical-Plating Techniques

Vacuum deposition of silver, gold, platinum, niobium, uranium, and rhenium films for use in diffusion studies at Harwell is described.²⁸ The induction-heated metals were satisfactorily evaporated from thoria crucibles at temperatures up to 2500°C.

The rate of deposition of niobium by thermal decomposition of niobium iodides was studied at Du Pont²⁹ in the evaluation of the iodide process for the preparation of pure niobium. Effects of deposition temperature and temperature of the

feed material on the rate of deposition were investigated.

U. S. Patent 2,894,320 (reference 30) describes the coating of uranium metal with nickel, molybdenum, chromium, niobium, and tungsten by thermal decomposition of the corresponding carbonyls. The bonding is improved by subsequent thermal diffusion of the coating.

Preparations were made at Atomics International³¹ for the deposition of molybdenum and chromium coatings on uranium-molybdenum fuel slugs by thermal decomposition of the corresponding metal carbonyls. A coating apparatus is described.

Problems in scaleup of fluidized-bed chemical reactors for the vapor deposition of metal coatings on nuclear-fuel particles have been considered.³² Proper operating conditions for optimum efficiency of the jet type reactor are predicted.

Single crystals of UO_2 , 6 to 10 mm in length, were prepared by hydrolysis of UF_4 at 1050°C .³³

An effort is being made to obtain the UC by the reaction of iodine-stabilized UI_4 with a hydrocarbon in a fluidized bed of seed particles.³³⁻³⁵ It is anticipated that this product would be coated with pyrolytic carbon, without interruption of the fluidization process, to give a useful fuel material which could be handled in air and whose coating may be sufficiently dense to retain fission fragments.

Ceramic Coatings

Battelle³⁵ is investigating sintered cladding for UO_2 fuel particles. Batches containing 55 to 75 wt.% UO_3 were studied. Particles in which the initial fuel composition was 65 wt.% or more UO_3 showed relatively low weight gains in air at 1200°F . With a UO_2 content of over 25 wt.%, the oxidation resistance fell off rapidly.

One method³⁴ of retaining fission products in a spherical graphite fuel element is to place a coating or seal between the fueled graphite and the unfueled graphite shell. The wetting and penetration of a few materials have been studied by heating in graphite crucibles at 1800, 2200, and 2600°F . Metallographic examination of the specimens after heating indicated that copper, nickel, silicon, and two barium-containing glasses may be suitable seal materials.

(B. W. King)

Explosive Forming

The ABC's of explosive forming have been outlined by Pearson³⁶ of the U. S. Naval Ordnance Test Station at China Lake, Calif. He states that the behavior of metals under explosive conditions depends primarily on the properties of the metal being formed and on the forming system that is employed. Much of the confusion that has evolved concerning explosive forming has occurred because of attempts to compare a wide range of metals and methods of operation. The method of operation employed in the forming of steel, for example, does not apply for forming zirconium because of the differences in mechanical properties. A better understanding of the behavior of explosively treated materials can be obtained by considering the material particle speed rather than over-all stresses. The limit of speed that particles can travel in a metal is called the critical relative particle velocity, and, when it is exceeded, the metal ruptures. Another factor that plays an important role in explosive forming is the control of the reflected waves. When an explosively generated compression wave (pulse) is transmitted through a sandwich of dissimilar metals (for example, steel on aluminum) it travels through the steel until it reaches the steel-aluminum interface, where about half of the generated energy will continue on through the aluminum, whereas the other half will bounce back into the steel and complicate the internal stress picture.

The effect of high-speed forming on the yield and ultimate strengths of some metals has been demonstrated. Speeding up the rate of forming increases the yield and ultimate strengths. However, the effect on the amount of elongation is not always consistent. Some metals show increases in elongation, whereas others show decreases.

Basic tools required in explosive forming consist of a female die of the required shape, which is vented to permit the escape of entrapped air at the die face during forming; an explosive; and a medium to transmit the shock wave generated by the explosive. Any one of these items can be varied to produce a very definite effect on the operation. Either low (deflagrating) or high (detonating) explosives can be used as the energy source. However, the use of high explosives has been more popular since a low explosive requires some type of enclosure to improve its efficiency. Three fac-

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U. S. Patent 2,894,320 (reference 30) describes the coating of uranium metal with nickel, molybdenum, chromium, niobium, and tungsten by thermal decomposition of the corresponding carbonyls. The bonding is improved by subsequent thermal diffusion of the coating.

Preparations were made at Atomic International³¹ for the deposition of molybdenum and chromium coatings on uranium-molybdenum fuel slugs by thermal decomposition of the corresponding metal carbonyls. A coating apparatus is described.

Problems in scaleup of fluidized-bed chemical reactors for the vapor deposition of metal coatings on nuclear-fuel particles have been considered.³² Proper operating conditions for optimum efficiency of the jet type reactor are predicted.

Single crystals of UO_2 , 6 to 10 mm in length, were prepared by hydrolysis of UF_4 at 1050°C .³³

An effort is being made to obtain the UC by the reaction of iodine-stabilized UI_4 with a hydrocarbon in a fluidized bed of seed particles.³³⁻³⁵ It is anticipated that this product would be coated with pyrolytic carbon, without interruption of the fluidization process, to give a useful fuel material which could be handled in air and whose coating may be sufficiently dense to retain fission fragments.

Ceramic Coatings

Battelle³⁵ is investigating sintered cladding for UO_2 fuel particles. Batches containing 55 to 75 wt.% UO_3 were studied. Particles in which the initial fuel composition was 65 wt.% or more UO_3 showed relatively low weight gains in air at 1200°F . With a UO_2 content of over 25 wt.%, the oxidation resistance fell off rapidly.

One method³⁴ of retaining fission products in a spherical graphite fuel element is to place a coating or seal between the fueled graphite and the unfueled graphite shell. The wetting and penetration of a few materials have been studied by heating in graphite crucibles at 1800, 2200, and 2600°F . Metallographic examination of the specimens after heating indicated that copper, nickel, silicon, and two barium-containing glasses may be suitable seal materials.

(B. W. King)

Explosive Forming

The ABC's of explosive forming have been outlined by Pearson³⁶ of the U. S. Naval Ordnance Test Station at China Lake, Calif. He states that the behavior of metals under explosive conditions depends primarily on the properties of the metal being formed and on the forming system that is employed. Much of the confusion that has evolved concerning explosive forming has occurred because of attempts to compare a wide range of metals and methods of operation. The method of operation employed in the forming of steel, for example, does not apply for forming zirconium because of the differences in mechanical properties. A better understanding of the behavior of explosively treated materials can be obtained by considering the material particle speed rather than over-all stresses. The limit of speed that particles can travel in a metal is called the critical relative particle velocity, and, when it is exceeded, the metal ruptures. Another factor that plays an important role in explosive forming is the control of the reflected waves. When an explosively generated compression wave (pulse) is transmitted through a sandwich of dissimilar metals (for example, steel on aluminum) it travels through the steel until it reaches the steel-aluminum interface, where about half of the generated energy will continue on through the aluminum, whereas the other half will bounce back into the steel and complicate the internal stress picture.

The effect of high-speed forming on the yield and ultimate strengths of some metals has been demonstrated. Speeding up the rate of forming increases the yield and ultimate strengths. However, the effect on the amount of elongation is not always consistent. Some metals show increases in elongation, whereas others show decreases.

Basic tools required in explosive forming consist of a female die of the required shape, which is vented to permit the escape of entrapped air at the die face during forming; an explosive; and a medium to transmit the shock wave generated by the explosive. Any one of these items can be varied to produce a very definite effect on the operation. Either low (deflagrating) or high (detonating) explosives can be used as the energy source. However, the use of high explosives has been more popular since a low explosive requires some type of enclosure to improve its efficiency. Three fac-

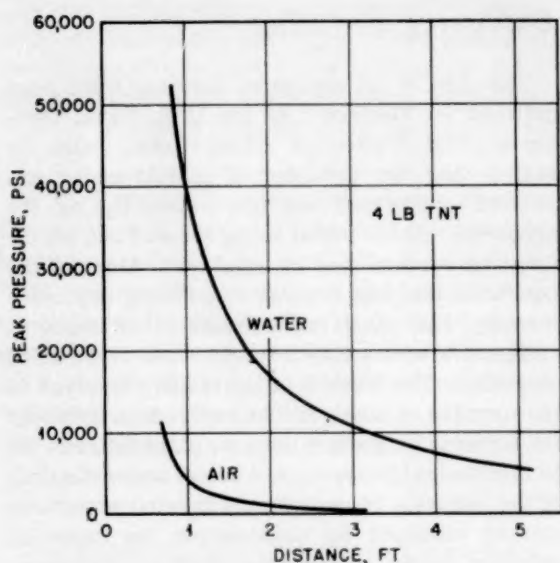
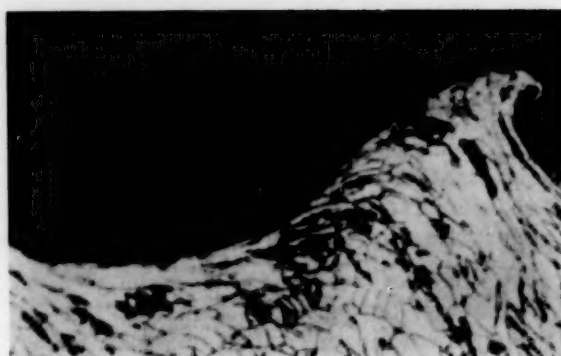


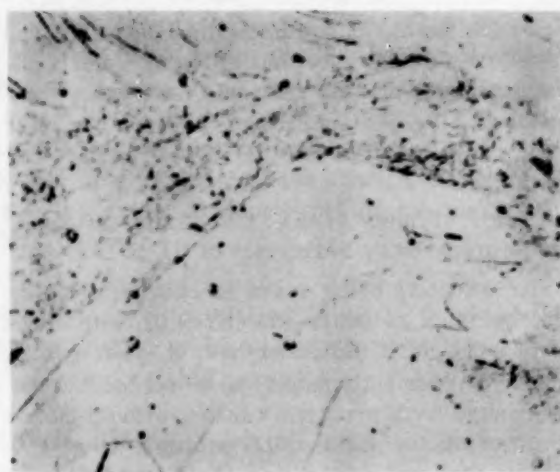
Figure 15—Effect of transfer medium on the generated peak pressure in explosion forming,³⁶ showing the amount of working force delivered by a fixed weight of charge in air and water at various distances from the workpiece. (Similar results are obtained when time is substituted for distance.) (Reprinted by permission from *Steel*.)

tors that influence the results obtained by using high explosives for a forming operation are (1) the type of explosive, (2) its geometric configuration, and (3) the distance it is mounted above the workpiece. The proper combination of these to produce optimum results must be experimentally determined for each forming operation and material.

Several transfer media, such as air, water, and semisolids, have been tested, and the type of transfer medium used depends primarily on the forming operation. Air has been used mainly for explosive welding and work hardening, whereas water has been used for deep drawing, embossing, shaping, and sizing. Water media are used for operations such as deep drawing because more work can be done with a given weight of charge, as shown in Fig. 15. In this case, the gases produced by the detonation of the explosive form a gas bubble that expands immediately with great force, creating a pressure wave in the surrounding water. This pressure wave, which is preceded by the shock wave, generates additional energy in the system which contributes to the forming of the workpiece. The Naval Ordnance Test Station has



(a)



(b)

Figure 16—Metallic jets and bond produced by explosive welding.³⁶ (a) Photomicrograph of the metallic jets produced by explosively driving two pieces of metal together. (b) Photomicrograph of the bond produced by explosive welding. Magnification, 100 \times . (Reprinted by permission from *Steel*.)

been conducting tests in explosive welding. Unsymmetrical loading and angles between metal pieces comprise the basic idea in explosive welding. Two pieces of metal moving rapidly toward each other with an included angle for collapse will produce metallic jets in the surface, as shown in Fig. 16. Too much speed displaces the tiny wavelike jets, whereas too little speed produces no interaction. The proper angle and speed produce jetting, interaction, and a permanent bond such as that shown in Fig. 16.

Borg-Warner³⁷ has conducted a program concerned with the effects of charge weight and standoff distance on the resulting bulge depth, as well as on the thickness, hardness, and yield

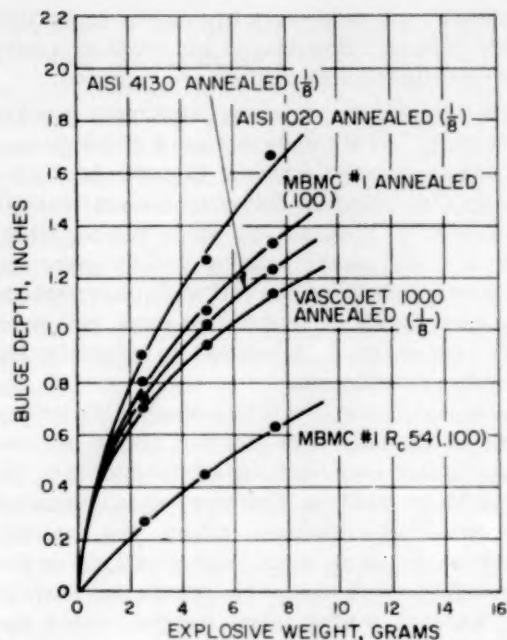


Figure 17—Relation between charge weight and bulge depth for various steels.³⁷

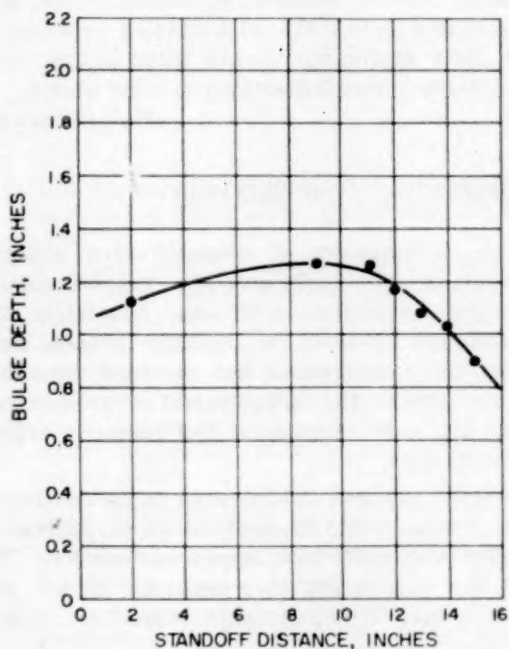


Figure 18—Relation between bulge depth and standoff distance for annealed AISI 1020 steel formed using a 5-g charge.³⁷

strength of the formed specimen. The materials used were AISI 4130, AISI 1020, MBMC No. 1, Vascojet 1000, and types 2024 and 6061 alu-

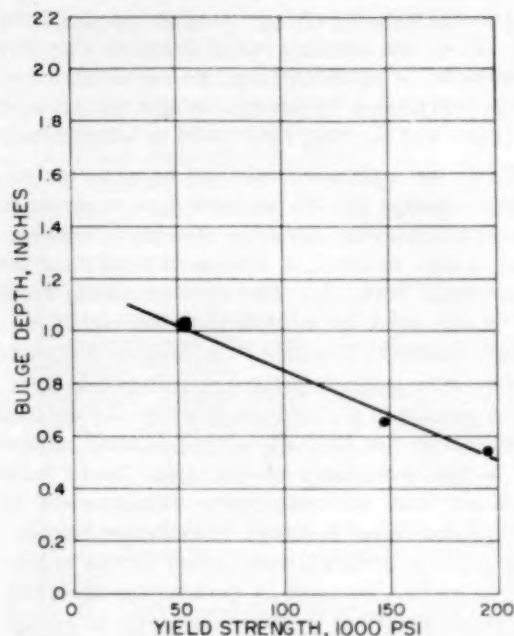


Figure 19—Relation between yield strength and bulge depth for AISI 4130 steel.³⁷ Conditions: 5 g at an 11-in. standoff distance.

minum. The testing procedure used consisted of placing the specimen in the tool set, filling the tool set with water, positioning the desired weight of charge at the predetermined standoff distance, and detonating. The explosive used for these tests was tetryl (trinitromethyl phenyl-nitramine), which was chosen because of its relative consistency of action.

The effect of the charge weight on the bulge depth for the various materials is shown in Fig. 17, and the effect of standoff distance on the bulge depth is shown in Fig. 18. The relation between yield strength and bulge depth for AISI 4130 is shown in Fig. 19. Similar relations were found to exist with the AISI 1020 and aluminum alloys tested. An attempt was also made during this program to correlate the bulge depth with elongation, hardness, and ultimate strength; however, no recognizable relation could be found. Data obtained in explosively forming an annealed AISI 4130 specimen indicated that no physical or structural changes occurred which would not have occurred during cold forming to a comparable configuration by conventional means.

The average metal-forming rate was also determined during this investigation. Annealed AISI 4130 specimens $\frac{1}{16}$ in. thick were used for

this work, and, for charge weights ranging from 7 to 10 g, the average metal-forming rate was calculated to be 297 ft/sec. No correlation between variations in charge weight or standoff distance and forming rate could be established.

Water as a transfer medium between the explosive charge and the workpiece is superior to air from the metal-forming viewpoint; however, when water is used, a means of insulating the tank walls from the stresses resulting from the blast must be considered. Insulating materials tested by Lockheed³⁸ included an inflated-rubber-tube liner, a pumice and aluminum liner, and a curtain of air bubbles generated by passing compressed air through a ring located around the bottom periphery of the tank. These tests revealed that air was quite effective as an energy-absorbing medium. The inflated-rubber-tube liner provided a substantial stress reduction; however, because of the expense involved, the most practical system appeared to be the aerator design. (C. C. Simons)

Welding and Brazing

Hanford²⁴ has initiated programs involving the use of zirconium-beryllium brazing alloys developed at Bettis and Armour. The nominal eutectic alloy has been used, but difficulties are apparent in controlling the composition. These difficulties lead to wide variations in the melting point of the alloy. For example, the melting point of powdered alloy ranges from 980 to 1030°C, depending on the mesh size of the powder. The Hanford results indicate that the eutectic alloy will be of limited usefulness unless composition is controlled very carefully. No recent mention of the zirconium-beryllium alloys with a ternary addition has appeared. A comparison of the sensitivity of the various binary and ternary alloys to the effects of compositional variation on melting point appears to be needed.

An eight-part catalog of reports on welding has been published which will be a useful tool for workers in the field.³⁹

In work conducted at Los Alamos on beryllium joining,⁴⁰ cracking in the weld or heat-affected zone was encountered in attempts to produce fusion welds without filler metal additions. The use of either pure silver, added during inert-gas welding with a tungsten electrode (TIG), or an aluminum-12 wt.% silicon alloy as the

electrode was necessary to produce acceptable welds. Specific procedures and conditions used in the configurations studies are reported.

The possibility of using aluminum powder metallurgy (APM) material as a cladding material has spurred interest in methods of producing end closures. Atomics International is conducting a program to study fusion, flash, spot, and ultrasonic welding and hot pressing as joining methods for APM.⁴¹ All methods have been fairly successful to date, but much more information is needed to evaluate the joints for reactor use.

A limited Battelle investigation⁸ of the weldability of high-strength niobium alloys has indicated resistance, spot, and TIG welding of the alloys to be feasible. However, when compared with other niobium-base alloys, the ductility of welds in these experimental alloys is not outstanding, and there is reason to suspect that thermal cycling might further reduce the ductility.

A brief review of the use of brazing for high-temperature service has been published by the Defense Metals Information Center.⁴² This review deals primarily with brazing processes and their application, braze filler alloys, and the effects of small additions to filler alloys.

(R. E. Monroe)

Nondestructive Testing

The development of nondestructive testing techniques for application to reactor-core-material inspection continues. Adaptation of established methods to specific shapes and materials combinations has received the bulk of the effort. The development of production tools and new techniques has received some attention also.

Several reports and reviews of nondestructive testing and its application to nuclear-reactor materials have appeared recently.⁴³⁻⁵⁰ For the most part, they describe the use of gamma and X-radiographic inspection techniques.

Ultrasonic echo and through-transmission techniques are being investigated for the inspection of both plates and tubes for flaws and inclusions.⁵¹ Experimentation at Hanford⁵² suggests the use of an ultrasonic technique for the measurement of plate thickness. The frequency of Lamb waves induced in the workpiece has

been found to be a sensitive measure of the plate thickness. An advantage claimed for this method is that the test head need not come in direct contact with the part being inspected. A test instrument is being developed.

An eddy-current technique for the measurement of the thickness of zirconium cladding on uranium and uranium oxide cores has been described by Du Pont.⁵³ A recently revised system is capable of measuring a zirconium thickness ranging from 6 to 15 mils with a linear response. The measurements are unaffected by scale or other surface conditions and are accurate even when the sensing head is raised as much as 4 mils from the surface. Hanford⁵⁴ continues to investigate the measurement of jacket thickness and jacket-to-core gap variations by eddy-current means.

The application of a thermographic technique in the measurement of core-to-cladding bond integrity⁵⁵ centers about the detection of temperature variations in the vicinity of poorly bonded areas by an infrared sensing device. The specimen is heated transiently by either eddy-current or inductive means.

(D. R. Grieser)

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